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# **Review of the Transport of Selected Radionuclides in the Interim Risk Assessment for the Radioactive Waste Management Complex, Waste Area Group 7 Operable Unit 7-13/14, Idaho National Engineering and Environmental Laboratory, Idaho**

## **Volume I**

USGS Scientific Investigations Report 2005-5026

U.S. Department of the Interior  
U.S. Geological Survey

# **Review of the Transport of Selected Radionuclides in the Interim Risk Assessment for the Radioactive Waste Management Complex, Waste Area Group 7 Operable Unit 7-13/14, Idaho National Engineering and Environmental Laboratory, Idaho**

## **Volume 1**

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### Conversion Factors

Multiply	By	To obtain
<b>Length</b>		
centimeter (cm)	0.3937	inch (in.)
millimeter (mm)	0.03937	inch (in.)
meter (m)	3.281	foot (ft)
kilometer (km)	0.6214	mile (mi)
micron ( $\mu\text{m}$ )	0.00003937	inch (in.)
<b>Area</b>		
square meter ( $\text{m}^2$ )	0.0002471	acre
square kilometer ( $\text{km}^2$ )	247.1	acre
square centimeter ( $\text{cm}^2$ )	0.001076	square foot ( $\text{ft}^2$ )
square meter ( $\text{m}^2$ )	10.76	square foot ( $\text{ft}^2$ )
square centimeter ( $\text{cm}^2$ )	0.1550	square inch ( $\text{ft}^2$ )
square kilometer ( $\text{km}^2$ )	0.3861	square mile ( $\text{mi}^2$ )
<b>Volume</b>		
cubic centimeter ( $\text{cm}^3$ )	0.06102	cubic inch ( $\text{in}^3$ )
liter (L)	61.02	cubic inch ( $\text{in}^3$ )
cubic meter ( $\text{m}^3$ )	35.31	cubic foot ( $\text{ft}^3$ )
cubic kilometer ( $\text{km}^3$ )	0.2399	cubic mile ( $\text{mi}^3$ )
<b>Flow rate</b>		
meter per day (m/d)	3.281	foot per day (ft/d)
meter per year (m/yr)	3.281	foot per year ft/yr)
millimeter per year (mm/yr)	0.03937	inch per year (in/yr)
<b>Mass</b>		
gram (g)	0.03527	ounce, avoirdupois (oz)
kilogram (kg)	2.205	pound avoirdupois (lb)
<b>Pressure</b>		
kilopascal (kPa)	0.009869	atmosphere, standard (atm)
kilopascal (kPa)	20.88	pound per square foot ( $\text{lb}/\text{ft}^2$ )
kilopascal (kPa)	0.1450	pound per square inch ( $\text{lb}/\text{ft}^2$ )

Density		
kilogram per cubic meter (kg/m <sup>3</sup> )	0.06242	pound per cubic foot (lb/ft <sup>3</sup> )
gram per cubic centimeter (g/cm <sup>3</sup> )	62.4220	pound per cubic foot (lb/ft <sup>3</sup> )
Radioactivity		
becquerel per liter (Bq/L)	27.027	picocurie per liter (pCi/L)
Specific capacity		
liter per second per meter [(L/s)/m]	4.831	gallon per minute per foot [(gal/min)/ft]
Hydraulic conductivity		
meter per day (m/d)	3.281	foot per day (ft/d)
Hydraulic gradient		
meter per kilometer (m/km)	5.27983	foot per mile (ft/mi)
Transmissivity*		
meter squared per day (m <sup>2</sup> /d)	10.76	foot squared per day (ft <sup>2</sup> /d)

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as follows: °F=(1.8×°C)+32

Temperature in degrees Fahrenheit (°F) may be converted to degrees Celsius (°C) as follows: °C=(°F-32)/1.8

Vertical coordinate information is referenced to the "North American Vertical Datum of 1988 (NAVD 88)"

Horizontal coordinate information is referenced to the "Idaho State Plane Coordinate System East Zone Datum of 1927 (NAD27)"

Altitude, as used in this report, refers to distance above the vertical datum.

Specific conductance is given in microsiemens per centimeter at 25 degrees Celsius (µS/cm at 25°C).

Concentrations of chemical constituents in water are given either in milligrams per liter (mg/L) or micrograms per liter (µg/L).

## Acronyms

<u>Acronym</u>	<u>Definition</u>
ANSI	American National Standards Institute
ANSTO	Australian Nuclear Science and Technology Organization
ASTM	American Society for Testing Materials
BLR	Big Lost River
BWIP	Basalt Waste Isolation Program
CEC	cation exchange capacity
COC	contaminant of concern
COPC	contaminant of potential concern
DO	dissolved oxygen
DOC	dissolved organic carbon
DOE	Department of Energy
DOT	Department of Transportation
DQO	data quality objective
EDS	electron microscopy/energy dispersive X-ray spectroscopy
EG&G	former site contractor
EPA	Environmental Protection Agency
EQ3/6	thermodynamic database
ERDA	Energy Research and Development Administration

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FSMs	field sampling methods
FSP	field sampling plan
FY	fiscal year
HATCHES	thermodynamic database
ICPP	Idaho Chemical Processing Plant
INEEL	Idaho National Engineering and Environmental Laboratory
INTEC	Idaho Nuclear Technology and Engineering Center
IRA	Interim Risk Assessment
IUPAC	International Union of Pure and Applied Chemistry
LEA	local equilibrium assumption
LMITCO	Lockheed Martin Idaho Technologies, Inc.
LSD	land surface datum
LSIT	Large Scale Infiltration Test
MCL	maximum concentration limit
MDL	minimum detection level
NEA	Nuclear Energy Agency
NIST	National Institute of Standards and Technology
OECD	Organization for Economic Cooperation and Development
PHREEQC	computer program (pH redox equilibrium equations in the C programming language)
PNNL	Pacific Northwest National Laboratory
QAP	quality assurance plan
QAPjP	quality assurance project plan
QC	quality control
RATEQ	biogeochemical reactive-transport model
RESL	Radiological and Environmental Sciences Laboratory
RI/FS	remedial investigation/feasibility study
ROD	record of decision
RWMC	Radioactive Waste Management Complex
SAP	sampling analysis plan
SCM	surface complexation model
SDA	Subsurface Disposal Area
SI	saturation indices
SMO	Sample Management Office
SOW	statement of work
SRP	Snake River Plain
TETRAD	multiphase flow and solute transport computer code
TDS	total dissolved solids
TOC	total organic carbon
TRA	Test Reactor Area
TSA	Transuranic Storage Area
USGS	United States Geological Survey
VOC	volatile organic compound
WAG-3	Waste Area Group 3
WAG-7	Waste Area Group 7
WRIT	Waste/Rock Interaction Technology
XRD	X-ray diffraction

## Chemical Symbols

Al	aluminum
Am	americium
AN-65	labradorite
Ba	barium
Br	bromide
Ca	calcium
CaCO <sub>3</sub>	calcium carbonate
CaCl <sub>2</sub>	calcium chloride
Ca(NO <sub>3</sub> ) <sub>2</sub>	calcium nitrate
Ce	cerium
Cl	chloride
Cm	curium
Co	cobalt
CO <sub>2</sub>	carbon dioxide
CO <sub>3</sub>	carbonate
Cs	cesium
DTPA	diethylenetriaminepentacetic acid
EDTA	ethylenediaminetetraacetic acid
Eh	redox potential
F	fluoride
fo-50/10-90	olivine
Fe	iron
HEDTA	N-hydroxyethylene-diaminetriacetic acid
H	hydrogen
H <sub>2</sub> O	water
HCl	hydrochloric acid
HCO <sub>3</sub>	bicarbonate
HClO <sub>4</sub>	perchloric acid
HNO <sub>3</sub>	nitric acid
K	potassium
N	nitrogen
Na	sodium
NaNO <sub>3</sub>	sodium nitrate
NH <sub>4</sub>	ammonium
NO <sub>2</sub>	nitrite
NO <sub>3</sub>	nitrate
Np	neptunium
NTA	nitrilotriacetic acid
Mg	magnesium
Mn	manganese
O <sub>2</sub>	oxygen
OH	hydroxide
P	phosphorus



$P_{CO_2}$	partial pressure of carbon dioxide
$PO_4$	phosphate
Pu	plutonium
Ru	ruthenium
Sb	antimony
$SiO_2$	silica
$SO_4$	sulfate
Sr	strontium
Th	thorium
U	uranium
pH	hydrogen ion activity
XO	metal oxide
Zn	zinc
$^2H$	hydrogen isotope
$^{18}O$	oxygen isotope
$\delta$	delta notation for oxygen and hydrogen stable isotope ratios

## **Symbols**

$q$	= flux density
$K_{sat}$	= saturated hydraulic conductivity
$\psi$	= water pressure
$\rho$	= density of water
$g$	= gravitational acceleration
$\theta$	= water content
$z$	= vertical distance
$K$	= hydraulic conductivity
$\partial$	= differential operator
$C$	= differential water capacity
$K_d$	= distribution coefficient (volume per mass)
$C_s$	= mass of solute adsorbed per mass of adsorbent
$C_{Aq}$	= mass of solute dissolved per volume of water
$R_F$	= retardation factor
$P_b$	= bulk density (mass per volume)
$K_r$	= distribution coefficient (mass per mass per unit volume or unit mass)
$S_T$	= surface site density
$M$	= molar concentration
$m$	= molal concentration
$K_a$	= distribution coefficient (defined on basis of surface area)
$S_A$	= specific surface area
$k$	= permeability

$k_v$  = vertical permeability  
 $k_h$  = horizontal permeability  
 $n_f$  = fracture porosity  
 $V_L$  = longitudinal dispersivity  
 $P_c$  = capillary pressure  
 $h$  = capillary rise  
 $\sigma$  = interfacial surface tension  
 $\Theta$  = contact angle  
 $b_f$  = fracture aperture  
 $k_{fc}$  = fracture continuum permeability  
 $V_T$  = transverse dispersivity  
 $I_g$  = gas-phase tortuosity  
 $n_m$  = effective matrix porosity  
 $^\circ$  = degrees  
 $\nabla$  = gradient  
 $\propto$  = proportionality



## **Abstract**

The U.S. Department of Energy (DOE) requested that the U.S. Geological Survey conduct an independent technical review of the Interim Risk Assessment (IRA) and Contaminant Screening for the Waste Area Group 7 (WAG-7) Remedial Investigation, the draft Addendum to the Work Plan for Operable Unit 7-13/14 WAG-7 comprehensive Remedial Investigation and Feasibility Study (RI/FS), and supporting documents that were prepared by Lockheed Martin Idaho Technologies, Inc.

The purpose of the technical review was to assess the data and geotechnical approaches that were used to estimate future risks associated with the release of the actinides americium, uranium, neptunium, and plutonium to the Snake River Plain aquifer from wastes buried in pits and trenches at the Subsurface Disposal Area (SDA). The SDA is located at the Radioactive Waste Management Complex in southeastern Idaho within the boundaries of the Idaho National Engineering and Environmental Laboratory. Radionuclides have been buried in pits and trenches at the SDA since 1957 and 1952, respectively. Burial of transuranic wastes was discontinued in 1982.

The five specific tasks associated with this review were defined in a "Proposed Scope of Work" prepared by the DOE, and a follow-up workshop held in June 1998. The specific tasks were (1) to review the radionuclide sampling data to determine how reliable and significant are the reported radionuclide detections and how reliable is the ongoing sampling program, (2) to assess the physical and chemical processes that logically can be invoked to explain true detections, (3) to determine if distribution coefficients that were used in the IRA are reliable and if they have been applied properly, (4) to determine if transport model predictions are technically sound, and (5) to identify issues needing resolution to determine technical adequacy of the risk assessment analysis, and what additional work is required to resolve those issues.

## Executive Summary

This review of the Interim Risk Assessment (IRA) for Waste Area Group 7 (WAG-7) 13/14 Operable Units focuses on the fate and transport of selected actinides—americium (Am), uranium (U), neptunium (Np), and plutonium (Pu)—in mixed transuranic wastes buried in the shallow subsurface at the Subsurface Disposal Area (SDA), Radioactive Waste Management Complex (RWMC), Idaho National Engineering and Environmental Laboratory. The purpose of the IRA is to quantify the risk to human health and safety associated with the potential release of toxic and hazardous waste buried in shallow pits and trenches in the SDA.

The review team believes that the principal investigators for the IRA have done a commendable job of integrating the available information, justifying the different approaches used, documenting their assumptions, and acknowledging data limitations that they view as critical to the conclusions reached in the assessment. The latter is especially noteworthy and appreciated by the review team because of the complex issues involved in this review. The review team also acknowledges the excellent cooperation of the principal investigators of the IRA study in responding to requests for clarification, additional information, and supporting documentation.

The review team shares many of the concerns about data limitations that were expressed by the principal investigators of the IRA and other supporting studies. It is premature to conclude that these data limitations invalidate the conclusions reached in the IRA; however, they do raise doubt about the overall rigor of the study.

The contaminant transport analysis that was used in support of the IRA is based on (1) a source-term release-rate model that assumes the entire inventory of buried actinides will be mobilized at different rates over an extended period of time, (2) laboratory- or literature-derived distribution coefficients ( $K_d$ s) that define the capacity of the media to remove contaminants that are presumed to be in solution or behave like they are in solution, and (3) a flow model that tracks the migration of waterborne contaminants in time and space. As pointed out in the IRA, the release-rate model has not been calibrated and remains a major deficiency that needs to be resolved before completion of the Comprehensive Remedial Investigation/Feasibility Study (RI/FS). Many of the factors that affect the fate and transport of contaminants in the subsurface apply as well to the release-rate model.

The review team did not address issues that involved definition of the source term and modeling of contaminant release rates, both of which add additional uncertainty to an already complex problem. It is clear that definition of the source term and release rates are at least as important as other issues related to the formulation of the computational scheme to predict actinide transport. The solubility of the actinides depends on their chemical form and on the chemistry of the water that comes into contact with the actinides. Water chemistry is affected by the degradation and dissolution of other wastes that are buried with or near the actinide-bearing wastes. The implications of local water chemistry on actinide solubility, release rates, and contaminant transport are uncertain.

With a few exceptions, predictions of future actinide concentrations (Am, U, Np, and Pu) in the Snake River Plain aquifer that are presented in the IRA are less than, and in many cases significantly less than, maximum contaminant levels established by the U.S. Environmental Protection Agency. Notable exceptions are U and Np. Predicted concentrations, however, are based on limited and incomplete data that, in the opinion of the review team, are not adequate to demonstrate adherence to the guiding philosophy for model development presented in the IRA.

The guiding philosophy in developing the numerical simulator (Magnuson and Sondrup, 1998) was to use technically defensible estimates for the  $K_d$ s and conservative estimates of model parameters wherever “realistic” estimates were not available. The intent of the IRA was to present a conservative (but not overly conservative or unrealistic) scenario. It was not the intent of the IRA to present a “worst-case scenario”. However, the distinction between what is conservative or technically defensible and what is realistic is oftentimes blurred.

The review team examined many supporting documents for the IRA in arriving at the conclusion that model results are not shown to be conservative because of incomplete and inadequate knowledge of subsurface flow conditions and transport processes. For example, the  $K_d$ s that were used for Am, U, and Pu transport were derived from a limited number of batch tests of a single composite sedimentary interbed sample. This sample was prepared from a mixture of five samples from different depths and stratigraphic units and sieved to represent only the fine fraction of an otherwise heterogeneous suite of samples with different physical and chemical properties. Forty percent of the bulk sample (the coarse fraction) was eliminated from the batch tests, possibly biasing the results toward higher  $K_d$ s because of the greater surface

area represented by the finer size fraction. Furthermore, the batch tests were conducted using synthetic water with concentrations of major cations and anions that were generally lower than those measured for ground water and perched water near the SDA. The lower ionic strength of the solution reduces competition for available sorbing sites leading to higher  $K_d$  estimates. The synthetic water also was undersaturated with respect to calcite, even though calcite is a common mineral phase in the sedimentary interbeds. This is a potentially serious departure from actual field conditions because aqueous stability and mobility of the actinides may be enhanced by the formation of weakly sorbing carbonate complexes.

The importance of carbonate complexation is demonstrated in the thermodynamic modeling that was conducted as part of this review. Formation of carbonate complexes was also suggested, along with colloidal transport, as possible explanations for the early elution of a "fast fraction" that was observed in the column experiments for Am and Pu referenced in this review. Although the  $K_d$ s selected for use in the numerical simulator were selected from the lower end of the range of these laboratory-derived measurements, it is not obvious that the limited data and experimental protocols provide adequate opportunity to demonstrate that these  $K_d$ s are reasonably conservative or technically defensible.

The  $K_d$  issue is further complicated by the manner in which  $K_d$ s are dealt with in the numerical simulator. The numerical simulator treats individual  $K_d$ s as constant and independent of the effects that other contaminants, both actinide and nonactinide, have on competition for sorbing sites. This approach assumes that the availability of sorbing sites is infinite. Additionally, model simulations of flow indicate that the downward flux of waterborne contaminants in the subsurface is not uniformly distributed. This is a realistic portrayal of infiltration and percolation in the unsaturated zone that places higher stresses locally on the sorption capacity of the media.

The review team is of the opinion that more, and presumably better, data are needed to justify predictions of actinide transport presented in the numerical simulator that forms the basis of the IRA. In the absence of "reliable field calibration opportunities" for the actinides, the reviewers believe that several of the simplifying assumptions and model parameters selected for use in the numerical simulator warrant much greater scrutiny than might otherwise be the case if it were possible to evaluate model performance on the basis of direct comparisons to field observations.

Calibration of the numerical simulator is based on matching short-term observations of perched water in the unsaturated zone (flow component) and short-term measurements of nitrate concentrations in the saturated zone (transport component). These calibration controls are only indirectly suggestive of the model's ability to predict long-term movement of the actinides. No attempt was made, and the review team believes rightly so, to calibrate the model to actinide detections in the field. For several reasons, calibration of actinide transport to actual field observations is not realistic. The limited number of detections, their sporadic and seemingly random nature, and uncertainty over whether or not these detections represent dissolved-phase transport, preclude their use for this purpose, and in that sense, we agree with the approach presented in the IRA. However, the validity of using the nondetects as general indicators of model performance is questionable and may even be misleading. The only valid conclusion that can be reached from the simulated results is that the model predicts actinide concentrations that are below the minimum detection level. Nothing can be said about whether the model overpredicts or underpredicts actinide concentrations because there is no way to objectively confirm or refute the claim.

Although the apparent lack of widespread contamination is good news, because it probably means that there is no wholesale release of the actinides at concentrations above the minimum detection level, it does present a dilemma. One of the best defenses of a model's ability to predict future outcomes is its ability to match the historic record. The longer the historic record, the higher the confidence in the predictions. To help mitigate the uncertainty that this dilemma presents, the review team feels that more rigorous definition of the hydrologic properties of the sedimentary interbeds and the actinide distribution coefficients associated with these geologic units are needed.

The numerical simulator for the IRA places considerable emphasis on the role of the sedimentary interbeds to arrest the movement of the actinides. The review team feels that this emphasis is appropriate. However, because the sedimentary interbeds figure so prominently in the modeling of flow and transport, thorough characterization of their hydrologic and geochemical properties should be a very high priority.

Characterization of the interbeds should include definition of the vertical and lateral variability of their physical, hydrological, and chemical properties. The validity of simplifying assumptions, that are needed to

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represent these geologic units in a numerical simulator, cannot be established without data that can show how these basic properties vary in space.

Similarly,  $K_d$ s for the sedimentary interbeds should be determined using samples that are truly representative of the different lithologic units and subunits present in the sedimentary interbeds. Homogenization of these samples, for purposes of expediting analysis or determining average properties, should be avoided. Batch and column  $K_d$  experiments should be conducted with solutions that more closely replicate both the natural- and contaminated-water chemistry in the vicinity of the RWMC. These tests should also include an assessment of the impact that other contaminants in solution have on measured  $K_d$ s. The validity of using a constant  $K_d$  for individual actinide species, needs to be evaluated in the context of a dynamically evolving chemical system. The evolution of this system is affected by temporal and spatial variations in water chemistry, actinide and nonactinide contaminant concentrations in the ground water, release rates of other contaminants that are buried at the SDA, and the affinity of these other contaminants to occupy sorption sites.

Finally, there is considerable uncertainty over how the exclusion of lateral flow of water from outside the SDA affects actinide predictions. Until recently (1999), there was very little evidence to demonstrate that water, diverted from the Big Lost River (BLR) into the spreading areas located west of the SDA, can migrate laterally in the unsaturated zone beneath the SDA. The conceptual model of flow in the unsaturated zone beneath the SDA assumes that essentially all flow is derived from precipitation and surface runoff that occurs within the immediate vicinity of the SDA. Exclusion of lateral flow from outside the SDA (BLR and/or spreading areas) may represent a significant departure from real world conditions. If so, then interpretations of field data that were used to calibrate the flow and transport component of the numerical simulator may be seriously compromised. Until the effects of lateral flow are better defined, then the reliability of the numerical simulator to predict the fate and transport of waterborne contaminants in the subsurface will remain controversial. Thus, the intent to present a "technically defensible" approach that would lead to a conservative (but not overly conservative or unrealistic) scenario has not been demonstrated.

## 1.0 Introduction

The Interim Risk Assessment (IRA) and Contaminant Screening for the Waste Area Group 7 (WAG-7) Remedial Investigation (Becker and others, 1998) was prepared for the U.S. Department of Energy (DOE) by Lockheed Martin Idaho Technologies, Inc. (LMITCO) as a formal record of work completed for the Waste Area Group Comprehensive Remedial Investigation and Feasibility Study (RI/FS). The draft Addendum to the Work Plan for Operable Unit 7-13/14 WAG-7 RI/FS (DOE, 1998) defined revised strategies and additional requirements for conducting the WAG-7 RI/FS. These two reports and supporting documents will be used to complete the draft Record of Decision (ROD) for remediation of WAG-7 by December 2002. The DOE, in order to prepare for ROD negotiations with the U.S. Environmental Protection Agency and the State of Idaho, requested that the USGS conduct an independent technical review of the Interim Risk Assessment, the Addendum, and associated documents.

The current version of the IRA represents a preliminary analysis of the potential risks to human health and safety associated with the release of buried mixed-chemical, low-level, and high-level wastes at the Radioactive Waste Management Complex (RWMC) Subsurface Disposal Area (SDA) (fig 1-1). The current IRA is a comprehensive evaluation of risks associated with all contaminants of potential concern (COPC's). This review of the IRA is concerned only with the analysis of predicted releases of long-lived actinides (Am, U, Np, and Pu) to the accessible environment. It is further limited to a review of the radionuclide data and geotechnical methodologies that were used in the numerical model to predict actinide migration in the subsurface. Evaluation of the contaminant source term and the model that was used to predict the rate and timing of releases of the actinides from buried waste containers are not included in this review. For purposes of this review, the accessible environment is defined as the Snake River Plain (SRP) aquifer.

The SDA comprises an area 975 m long from east to west and 520 m wide along its eastern boundary. Radioactive wastes have been buried within its boundaries in trenches since 1952 and in pits since 1957. The pits and trenches range from 15 to 91 m wide, 76 to 335 m long, and 1.4 to 4.6 m deep. Burial of transuranic wastes at the SDA ceased in 1982.

The potential for future releases of the actinides to the SRP aquifer and the risks associated with those releases depend on the source term (quantity and types of other contaminants present, their chemical form, and their

release rates) and the mobility of the actinides themselves. Actinide mobility presumably is enhanced by the presence of water because water moves and is constantly replenished, which sustains downward movement of the contaminant. The form of the contaminant determines how mobile it will be as an aqueous species, whether in true solution or in suspension. Its movement from beneath present burial sites to the accessible environment is controlled by water and rock interactions that are determined by the geology and chemistry of the environment that it comes into contact with. These complex physical and chemical processes can be expected to vary in both time and space.

### 1.1 Purpose

The purpose of this technical review was to evaluate the data and the geotechnical approaches that were used by LMITCO to estimate future risks associated with the release of four actinides (Am, U, Np, and Pu) to the accessible environment (SRP aquifer) from wastes buried in pits and trenches at the SDA.

### 1.2 Scope

The scope of this review and the organization of this report are defined by a series of five tasks, each framed as a question. These questions were developed from discussions with DOE based on a preliminary "Proposed Scope of Work" prepared by DOE and a follow-up workshop in June 1998 that was attended by review-team members, DOE, and LMITCO personnel. The workshop included summary presentations by the principal investigators from LMITCO who were involved in the development of the IRA and supporting studies, and a visit to the RWMC that was hosted by the DOE and LMITCO personnel involved in site monitoring activities.

The scope and objectives of the individual tasks identified in the review proposal are summarized as follows:

#### 1.2.1 Task 1: Review of radionuclide sampling data at the Radioactive Waste Management Complex

Question: How reliable and significant are the reported radionuclide detections, and how reliable is the ongoing sampling program?

The objective of Task 1 was to evaluate the reliability and significance of the reported detections of actinide radionuclides at the RWMC and to assess the reliability of the ongoing sampling program. The radionuclide species to be considered were Am, U, Np, and Pu. Data to be con-



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sidered were data presented in the IRA, data published by the USGS, and any additional data provided by LMITCO and DOE.

Multiple criteria were to be considered in evaluating the available data: (1) statistical significance, (2) isotopic association, (3) consistency of detections with the historical data record, (4) the possibility of cross contamination, and (5) the adequacy of the sampling program.

### 1.2.2 Task 2: Radionuclide transport processes

Question: What physical and chemical processes can logically be invoked to explain (true) detections?

The objective of Task 2 was to provide reasonable explanations to account for the observed transport of the actinide radionuclides at the SDA, as interpreted from data evaluated in Task 1. Both geochemical and hydrologic factors were to be considered in explaining true detections that were summarized in Task 1. The scope of this task involved (1) speciation calculations to evaluate what conditions at the SDA are consistent with enhanced mobility of Am, U, Np, and Pu, (2) an assessment of the role of colloid-facilitated transport on actinide movement in the subsurface, and (3) an evaluation of the possible role of preferential flow on actinide migration.

### 1.2.3 Task 3: Distribution coefficients ( $K_d$ s) and their application to transport analysis

Question: Are  $K_d$ s that were used in the IRA reliable and have they been applied appropriately?

The objective of Task 3 was to evaluate the reliability and appropriateness of  $K_d$ s that were used in the fate-and-transport modeling of the actinides as described in the IRA. The scope of this task involved (1) a review of the literature on  $K_d$ s that may be considered applicable to the RWMC and  $K_d$ s reported for similar subsurface sediments, (2) an evaluation of  $K_d$ s used in the IRA in terms of the existing subsurface mineralogy and aqueous geochemical conditions, (3) an evaluation of the experimental protocols used to determine  $K_d$ s, (4) determination of the extent to which the  $K_d$  concept may be applicable to the site, and (5) an assessment of the uncertainty associated with the  $K_d$ s used for fate-and-transport modeling.

### 1.2.4 Task 4: Transport model analysis

Question: Are model predictions defensible?

The objective of Task 4 was to evaluate whether or not the 3D model predictions of actinide transport made in support of the IRA are defensible. The scope of this task

includes (1) an evaluation of the conceptual model, (2) and assessment of the numerical model's representation of the stratigraphic framework of the fractured basalts and the role of the sedimentary interbeds, (3) an evaluation of the adequacy of the data sets selected for model calibration, and (4) an assessment of the predictive capabilities of the model.

### 1.2.5 Task 5: Further work

Question: What are the most significant issues that need to be resolved to support and defend the risk assessment analysis?

Question: What additional work is required to resolve those issues?

The objective of Task 5 was to provide timely interim guidance to several questions. First, what are the most significant issues that need to be resolved to support and defend the risk assessment analysis? Second, what additional work is immediately required to resolve those issues? This interim guidance was offered to provide sufficient lead time to resolve those issues within the time frame of the draft ROD (2002).

The scope of Task 5 included preliminary reviews of the Interim risk Assessment (Becker and others, 1998), results of the Large-Scale Infiltration Test (Wood and Norrell, 1996), Distribution Coefficients and Contaminant solubilities for the Waste Area Group 7 Baseline Risk Assessment (Dicke, 1997), and the flow and transport model (Magnuson and Sondrup, 1998). Additionally, planned work items were evaluated that were identified in the Addendum to the Work Plan (DOE/ID, March 1998). The intent of this task was to focus primarily on data needs rather than analytical approaches.

## 1.3 Acknowledgments

Preparing a report of this magnitude requires the support of many people. The authors acknowledge the contributions of the following USGS personnel: Elizabeth Jones compiled solid-phase data contained in chapter 3, and the solid-phase data tables were verified against the original data tables by Betty J. Tucker. The authors appreciate the technical reviews of the report that were conducted by LMITCO employees Laurence C. Hull, A. Jeffrey Sondrup, Swen O. Magnuson, Carol Craiglow, and Bruce H. Becker.

This report was prepared by the U.S. Geological Survey in cooperation with the U.S. Department of Energy's Idaho Operations Office (DOE). Kathleen E. Hain (DOE) coordinated the flow of information between the USGS and LMITCO.

## **1.4 Report Organization**

This report is organized into 7 chapters—Introduction, Conceptual Model, and Tasks 1 through 5. Each chapter with the exception of the introduction contains its own summary.

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## 2.0 Conceptual model of aqueous-phase fluid flow and contaminant transport

### 2.1 Regional setting

The Subsurface Disposal Area (SDA) (fig. 1-1) is on the eastern Snake River Plain (SRP), and the ground water immediately below it is part of the large and heavily used SRP aquifer. The climate is semiarid; average annual precipitation is about 22 cm. Subsurface water movement is affected by a variety of topographic and hydrogeologic features, both natural and artificial. The unsaturated zone at the SDA is unusually thick—about 200 m. It also is complex, comprising granular media and consolidated, fractured rock, both of which affect the subsurface hydrology in many ways. These media are highly stratified. The contrasts in properties of adjacent layers may impede downward flow but also may lead to various types of fast, or preferential flow. Near the SDA, surface-water features, notably the Big Lost River (BLR) and spreading areas, sometimes are major sources of infiltration.

Key issues related to subsurface contaminant transport include (1) travel times to and within the aquifer, both average or typical values and the range of values to be expected, and (2) modes of contaminant transport, especially adsorption processes. Some of the complicating factors are the effectiveness of natural and artificial barriers; the direction (horizontal, vertical, or other) of flowpaths; the diffuse or preferential nature of flow; the chemical nature of the contaminants and subsurface media; and sources of water now in the subsurface, such as local precipitation, runoff, and lateral flow from spreading areas or elsewhere.

### 2.2 Conceptual models

In hydrologic contexts, the term “conceptual model” is used in at least two different ways: (1) An ideal sort of conceptual model was defined by Hoxie (1989) with a quote from Russell (1948) that considers a conceptual model of a natural system to be a “hypothesis which fits the (available) data, which are as simple as compatible with this requirement, and which make it possible to draw inferences subsequently confirmed by observation,” and (2) An applied conceptual model was defined by Anderson and Woessner (1992) as “a pictorial representation of the ground water flow system, frequently in the form of a block diagram or a cross section. The nature of the conceptual model will determine the dimensions of the numerical model and the design of the grid.” In both

usages, the conceptual model relates a physical system to a mathematical model, though definition 1 emphasizes the validity of the connection to the physical system, requiring it to produce “inferences subsequently confirmed by observations,” whereas definition 2 is less rigorous, emphasizing the practical connection to the mathematical model, but not implying that the predictions will necessarily be realistic. In USGS research at Yucca Mountain in Nevada, there has been a continual effort, presented in a series of publications (Montazer and Wilson, 1984; Wittwer and others, 1992; Hoxie, 1989), to create and develop a conceptual model that is intended, with each refinement, to more closely approximate an ideal conceptual model. Research at the INEEL SDA relies heavily on a practical conceptual model that has not been explicitly presented in the INEEL reports but is implied by the assumptions and other details of numerical modeling (Becker and others, 1998; Magnuson and Sondrup, 1998). In this chapter, we describe our best approximation of an ideal conceptual model of transport processes at the SDA. In chapter 6, we explicitly evaluate the practical conceptual model used in the most advanced numerical modeling of actinide transport at this site.

The construction of an ideal conceptual model necessitates choice among competing hypotheses, and recognition of uncertainties of individual components of the model. One example of competing hypotheses is whether sedimentary interbeds are a major retarding influence on vertical contaminant transport or an expediting influence because of preferential flowpaths within them. Another is whether the perched water commonly observed within and above interbeds comes from local infiltration whose downward flow is impeded or from lateral flow from areas that sometimes are sources of substantial infiltration (for example, the spreading areas west of the SDA).

### 2.3 Geologic framework

The geologic framework of the SDA consists of interbedded basalts and sediments of Quaternary age. Basalt flows erupted from numerous fissures and small shield volcanoes located on and near the Arco-Big Southern Butte volcanic rift zone (Kuntz and others, 1992). Sediment consists of alluvial and eolian deposits derived from the ancestral channel and floodplain of the BLR (Rightmire and Lewis, 1987b; Hughes, 1993). These deposits make up a thick unsaturated zone and the SRP aquifer at the SDA.

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### 2.3.1 Geologic data

The geologic framework of the SDA and adjacent areas has been characterized on the basis of data from 94 wells (Magnuson and Sondrup, 1998). About half of these wells are at and near the SDA (figs. 2-1 and 2-2). Continuous cores ranging in length from less than 30 m to 550 m were obtained from 34 of the 94 wells (Davis and others, 1997); however, only three of these cores are longer than 90 m. Most basalt cores represent nearly complete recovery. Past recoveries of sediment cores ranged from 0 to 100 percent, but commonly were less than 50 percent (Hughes, 1993; Burgess and others, 1994). Common borehole geophysical logs, such as caliper, neutron, natural-gamma, and density logs, were obtained from most wells (Bartholomay, 1990a). These wells include 20 U.S. Geological Survey wells that are labeled as USGS 9, 87, 88, 89, 90, 91, 92, 93, 93A, 94, 95, 96, 96A, 96B, 105, 109, 117, 118, 119, and 120 in figures 2-1 and 2-2.

Numerous studies have been conducted to characterize the geohydrologic properties of basalt and sediment at the SDA. Kuntz and others (1980) described the geologic ages, petrographic characteristics, and paleomagnetic properties of basalt cores from wells 77-1, 76-6, USGS 93A, USGS 94, and USGS 95, located at and near the SDA (fig. 2-2), that form the foundation of stratigraphic interpretations at the SDA; basalt cores from three additional wells, C1A, WWW 1, and USGS 118, were evaluated for paleomagnetic properties at a later time. Stratigraphic relations were refined by Anderson and Lewis (1989) and Anderson and others (1996) using a combination of these core data and natural-gamma logs from 72 wells at and near the SDA. Rightmire (1984) and Rightmire and Lewis (1987b) described the grain-size distributions, bulk mineralogy, clay mineralogy, carbonate contents, and cation exchange capacities of surficial sediment and sedimentary interbeds on the basis of samples from 18 coreholes and selected waste pits at and near the SDA. Hughes (1993) described the mean grain sizes, sorting, skewness, kurtosis, general lithologies, and interpreted depositional environments and sedimentary structures of interbeds on the basis of samples obtained from 17 shallow cores at the SDA. Hydraulic characteristics of the basalt and sediment have been evaluated on the basis of numerous aquifer tests (Ackerman, 1991; Wylie and others, 1995), *in situ* and laboratory measurements of geologic materials (Bishop, 1991; Bishop, 1996; Welhan and Wylie, 1997), and simulations of data obtained from the Large-Scale Infiltration Test (LSIT), which was conducted about 1.5 km south of the SDA (Magnuson, 1995). Despite these and other studies, currently not enough is known about the characteristics and distribution of the

fine-scale features of basalt and sediment that are most likely to affect the potential for downward and lateral movement of water and wastes at the SDA. This is because these features throughout the SDA have not been characterized in a systematic and comprehensive way.

### 2.3.2 Stratigraphy

At least 11 basalt-flow groups, 10 sedimentary interbeds, and a veneer of surficial sediment are present between the land surface and the uppermost 60 m of the aquifer at the SDA (fig. 2-1) (Anderson and Lewis, 1989; Anderson and Bartholomay, 1995; Anderson and others, 1996). Basalt-flow groups informally are referred to as A through I. Sedimentary interbeds are referred to as A-B through H-I; interbeds A-B, B-C, and C-D are the thickest and most widespread interbeds. They also have been referred to in previous studies as the 30-ft (9-m), 110-ft (34-m), and 240-ft (73-m) interbeds, respectively. In this report, the letter designation will be used to identify these interbeds. Each flow group consists of from one to five separate basalt flows (Kuntz and others, 1980). Sedimentary interbeds and the surficial sediment consist of well sorted to poorly sorted deposits of clay, silt, sand, and gravel (Rightmire and Lewis, 1987a, b; Hughes, 1993). Basalt makes up about 90 percent of the volume of this stratigraphic section. Geologic ages of basalt flows in the section range from about 100 thousand years for those of flow group A to about 640 thousand years for those of flow group I. Ages of sedimentary interbeds A-B, B-C, and C-D range from about 100 thousand to 230 thousand years. The surficial sediment has been deposited during the past 100 thousand years following the eruption of basalt-flow group A. Because of their young ages and shallow depths, the surficial sediment and sedimentary interbeds A-B, B-C, and C-D are mostly unconsolidated.

### 2.3.3 Basalt flows

Source vents for most basalt flows at and near the INEEL, including those at the SDA, are concentrated in volcanic rift zones that trend perpendicular to the axis of the eastern SRP and parallel to the adjacent mountain ranges (Kuntz and others, 1992; Kuntz and others, 1994). Volcanic rift zones are characterized by eruptive and non-eruptive fissures, dikes, monoclines, faults, graben, and volcanoes having elongated slot-shaped vents (Rodgers and others, 1990; Kuntz, 1992; Kuntz and others, 1992; Smith and others, 1996). The distribution of fissures, dikes, and volcanoes is of hydrologic importance at and near the SDA because these features probably are numer-

ous and may greatly affect the range and distribution of hydraulic conductivity and the movement of ground water and wastes (Anderson and others, 1999). Areas proximal to volcanic vents are composed of highly permeable basalt flows and other volcanic deposits that may provide localized, preferential pathways for ground-water flow and movement of wastes. Dikes are thin, dense, vertical sheets of intrusive rock that locally may impede the movement of ground water and wastes. Noneruptive fissures that parallel dikes locally may provide additional highly permeable conduits for ground-water flow and movement of wastes. The SDA lies within a suspected vent corridor, one of many narrow, northwest-trending extensional features within and near the Arco-Big Southern Butte volcanic rift zone where there is a high probability for concealed volcanic vents, feeder dikes, and open fissures (Anderson and Liszewski, 1997; Anderson and others, 1999). The heterogeneity of volcanic rocks within this and other nearby vent corridors may impart complex anisotropy to the hydrologic system at and near the SDA.

### 23.3.1 Typical basalt-flow characteristics

A basalt-flow group, such as A through I at the SDA (fig. 2-1), consists of one or more distinct basalt flows deposited during a single, brief, eruptive event (Kuntz and others, 1980). All basalt flows of a group erupted from the same vent or several nearby vents, represent the accumulation of one or more lava fields from the same magma, and have similar geologic ages, paleomagnetic properties, and chemical compositions (Anderson and Bartholomay, 1995). The basalt flows mostly were deposited as tube-fed pahoehoe flows in the medial to distal parts of their respective lava fields. Individual flows generally range from 3 to 15 m thick and locally are interbedded with scoria and thin layers of sediment. Tube-fed pahoehoe flows, which are the most common type of flows in the eastern SRP, are characterized by dense interiors and rubbly, vesicular tops and bottoms cut by horizontal and vertical cooling fractures. Fractures and vesicles commonly are coated with fine-grained sediment infill and sometimes with secondary minerals consisting of calcite, clays, and zeolites (Rightmire and Lewis, 1987b; Morse and McCurry, 1997). Fractures within and contacts between individual basalt flows provide a complex network of potential vertical and horizontal pathways for the movement of water and wastes within the unsaturated zone and the aquifer.

### 23.3.2 Typical basalt-flow geochemistry

Most basalt flows at the INEEL have the chemical characteristics of both tholeiitic and alkali olivine basalts.

The small silica and large iron contents indicate that liquid temperatures were approximately 1,050 °C (Stout and Nichols, 1977). These basalts generally are medium to dark gray and range from vesicular, having elongated vesicles up to 4 cm in length, to dense. Typical basalt samples consist mainly of plagioclase feldspar (averaging An<sub>65</sub>, the composition of labradorite), pyroxene (tentatively identified as augite), and olivine (Fo<sub>50</sub> to Fo<sub>90</sub>); and contain lesser amounts of ilmenite, magnetite, hematite, and accessory apatite (Kuntz and others, 1980; Rightmire and Lewis, 1987b; Knobel and others, 1997). Chemical compositions of selected basalt samples at the INEEL were presented in reports by Kuntz and Dalrymple (1979), Knobel and others (1995), Reed and others (1997), and Collelo and others (1998). Stout and Nichols (1977) state that the augite contains 18.8 percent CaO and that opaque minerals constitute between 7 and 21 percent of the rock. With the exception of some basalt flows at Craters of the Moon (about 2 to 18 thousand years old) and basalt flows associated with Cedar Butte (about 420 thousand years old), there are no significant differences in chemical or mineralogical composition related to age or geographic location among basalt flows of the eastern SRP.

### 23.3.3 Fractures

Basalt flows on the eastern Snake River Plain and underlying the SDA contain abundant horizontal and vertical cooling fractures. Many of these fractures are interconnected and provide potential pathways for the movement of water, suspended sediment, and wastes in the subsurface. In general, the number of and aperture widths of fractures are much greater near the top and bottom of a basalt flow and are greatest along the top surface. Knutson and others (1992) describe a range of typical aperture widths measured from outcrops at the INEEL of from 0.0005 to 0.0025 m; however, aperture widths may range from several micrometers to several meters (Rightmire and Lewis, 1987b). Some of the widest, longest, and deepest fractures occur along the edges of sinuous lobes of inflated pahoehoe flows.

Rightmire and Lewis (1987a) describe fractures in basalt core samples from the SDA that range from those having fresh surfaces to those containing abundant sediment infill. Fractures having fresh surfaces most likely are dead-end fractures that transmit little, if any, water (Wood and Norrell, 1996; Magnuson and Sondrup, 1998). Fractures containing sediment infill or coatings probably are interconnected fractures that periodically transmit large quantities of water. Sediment in these fractures ranges from clay- to sand-sized and was transported by water and wind mainly during the deposition of overlying sedimen-

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tary interbeds. Sediment relations indicate multiple episodes of fracture filling during the geologic past. Post-depositional fracture filling probably continues as infiltrating water transports sediment. Sediment coloration indicates a wide range of minerals and depositional environments and includes hues of tan, brown, red, gray, orange, yellow, and green. Many of the deposits are calcareous. Additional fracture coatings consist of amorphous silica, precipitates consisting of calcite and mixed-layer illite/smectite clays, and possibly zeolites (Rightmire and Lewis, 1987a).

### 2.3.4 Surficial sediment and sedimentary interbeds

The geologic framework of the SDA has been affected by periodic eruptions of basalt followed by long periods of basin subsidence and sediment accumulation. The surficial sediment and sediment from the A-B, B-C, and C-D interbeds most likely represent deposition from meltwater discharge and periodic floods along the ancestral channel and floodplain of the BLR during past glacial declines (Hughes, 1993; Rathburn, 1993). The presence of gravel in the surficial sediment and the aforementioned interbeds indicates that a watercourse with flow capable of moving gravel at least occasionally traversed the area of the SDA during the past few hundred thousand years. Loess, windblown sediment having grain sizes ranging from fine silt to very fine sand, is a major part of the surficial sediment and sedimentary interbed A-B and, although not documented, probably also is present in sedimentary interbeds B-C and C-D.

The SDA is located near the southern end of the Big Lost trough (Gianniny and others, 1997), an area of past sediment accumulation along the channel and floodplain of the BLR and between the BLR sinks and Mud Lake (fig. 1-1). Sediment within this trough generally grades from a predominance of sand and gravel near the SDA to mainly clay and silt near Mud Lake. Barraclough and others (1976) hypothesized that the SDA was in the floodplain of the BLR prior to the Quaking Aspen Butte eruption and deposition of basalt-flow group A about 100 thousand years ago (Kuntz and others, 1994). Similar migrations of the BLR channel and floodplain, into and away from the SDA, probably occurred many times during the geologic past in response to the eruptions of basalt-flow groups B through I. During periods of volcanic quiescence, sediment accumulated in the topographic depressions of underlying basalt flows. Eolian material derived from the finer fraction of alluvial deposits probably accumulated in these and other areas, including along the flanks of volcanoes. Sedimentary units having large

amounts of clay-sized material probably were deposited in small lakes, some of which were formed by lava dams. Clay- to sand-sized sediment also was transported by water and wind into the fractures of underlying basalt flows. The thicknesses and areal extents of interbeds A-B, B-C, and C-D suggest that these units, like the surficial sediment, accumulated for long enough periods of time to fill and overtop most local topographic depressions. Sediment did not accumulate on some local basalt ridges, and sediment accumulation of interbed A-B was restricted in areal extent by the northward-sloping surface of basalt-flow group B, which erupted from a vent south of the SDA, near Big Southern Butte (fig. 1-1). These kinds of gaps in interbeds may provide local pathways for rapid infiltration of water and wastes through basalt fractures in the unsaturated zone.

#### 2.3.4.1 Depositional environments

Although the surficial sediment at the SDA contains a few minor sand-and-gravel lenses it consists predominantly of clay- and silt-sized material deposited as loess (Rightmire and Lewis, 1987a, b). Two periods of loess accumulation in this surficial sediment have been identified: one from about 80 to 60 thousand years ago, and one from about 40 to 10 thousand years ago (Forman and others, 1993). Two different interpretations have been made regarding the depositional environment of sedimentary interbed A-B. According to McElroy and others (1989), the primary source of this sediment is the accumulation of loess. According to Hughes (1993), the sediment of interbed A-B consists mainly of very fine sand and silt and represents deposition in a floodplain environment. At least one period of loess accumulation in interbed A-B has been identified (Forman and others, 1993); the age of this loess is not precisely known, but it probably accumulated from about 150 to 140 thousand years ago. The sediment of interbed B-C consists mainly of sand and gravel that is interpreted to have been deposited in a braidplain setting in channel systems as wide as 300 m between topographic highs in the basalt (Hughes, 1993). The sediment of interbed C-D consists mainly of sand and silt that is interpreted to have been deposited in low-energy channels and floodplains (Hughes, 1993). The continuous nature of this interbed indicates deposition in a broad, shallow braidplain setting that aggraded to above most of the topographic highs in the basalt (Hughes, 1993). Although current (1999) interpretations of depositional environments are useful for evaluating overall sediment relations, they are still too generalized to be useful for evaluating the potential for movement of water and wastes.

### 2.3.4.2 Lithologic variations

Significant lithologic variations occur within the surficial sediment and sedimentary interbeds A-B, B-C, and C-D owing to differences in depositional environments through time (Rightmire and Lewis, 1987a, b; Hughes, 1993). The lithology of interbed F-G and other deep interbeds has not been characterized. Fine-scale (centimeter) vertical and horizontal lithologic variations within the surficial sediment and sedimentary interbeds A-B, B-C, and C-D beneath the SDA generally are poorly defined. This is because too few sediment cores have been described in sufficient detail to quantify these variations. The most detailed descriptions of vertical lithologic variations were reported by Rightmire and Lewis (1987a) for coreholes 76-1, 76-2, 76-3, 76-4, 76-4A, 76-5, 76-6, and 77-2 (fig. 2-2). Samples from these coreholes include many thin, discrete layers or lenses of plastic clay, especially within the C-D interbed. The most detailed descriptions of horizontal lithologic variations were reported by Hughes (1993), but these descriptions are somewhat generalized. Both Rightmire and Lewis (1987a) and Hughes (1993) examined the sedimentary materials from coreholes 76-1, 76-2, 76-3, 76-4, 76-4A, 76-5, and 76-6, but there are many discrepancies between lithologic descriptions in these independent studies. For example, Rightmire and Lewis (1987a) described abundant plastic clay in the lower part of the C-D interbed from corehole 76-5, whereas Hughes (1993) described this same interval as sandy silt and slightly silty, fine to coarse sand. These discrepancies and the paucity of detailed descriptions of sediment cores make it difficult to quantify the role of these sedimentary materials in unsaturated-zone processes.

### 2.3.4.3 Physical characteristics and mineralogy

Other factors, such as sedimentary structures, grain-size distributions, bulk mineralogy, clay mineralogy, and ion exchange capacity may retard or enhance the movement of wastes through the surficial sediment and sedimentary interbeds A-B, B-C, and C-D. However, like lithology, these features have not been characterized in a systematic and comprehensive way across the SDA. Sedimentary structures include paleosols, cracks formed by hydrocompaction and dessication, freeze-thaw features, burrow and rootlet holes, caliche development, horizontal laminations, ripple cross-stratification, planar cross-stratification, lenticular bedding, flaser bedding, rip-up clasts, load casts, and varves (Rightmire and Lewis, 1987b; Hughes, 1993). Many sedimentary materials are dark reddish-brown which may result from dehydration and oxidation of iron-rich minerals by heat from the overlying lava flows. This coloration also may result from oxidation

of the ferrous iron to ferric iron in a historical oxygenated soil atmosphere where ferrous iron is released during the weathering of olivine and augite (Rightmire and Lewis, 1987b). Grain-size distributions, determined on the basis of sieve analyses of at least 236 samples from the surficial sediment and sedimentary interbeds A-B, B-C, and C-D, range from clay to pebble sized (Barraclough and others, 1976; Rightmire and Lewis, 1987b; Hughes, 1993). The median size of the grains in 46 samples analyzed by Barraclough and others (1976) was 0.066 mm, but the grain size probably was somewhat biased because of difficulty in recovering coarse sand and gravel lenses from interbeds while drilling. Fracture- and vesicle-infill sediments generally are finer grained than interbed sediments. This is expected because infiltrating water preferentially deposits finer grained particles in the vesicles and fractures (Rightmire, 1984).

Analyses of bulk mineralogy of sediments show the presence of quartz, plagioclase feldspar, potassium feldspar, pyroxene, olivine, calcite, and total clay minerals. Bartholomay (1990c, table 3, p. 11) statistically summarized the previously published bulk mineralogy data by interbed depth. Additional data on two cores from the SDA were published in a report by Reed and Bartholomay (1994). In general, quartz and plagioclase feldspar are the most abundant minerals in the sediment. Pyroxene and total clay minerals also are present in most of the sediment samples from the SDA. Abundance of clay minerals in interbed samples from the SDA ranges from 0 to 60 percent and averages about 20 percent (Bartholomay, 1990b). Olivine is present in some samples in trace amounts. Calcite also was identified in some samples. The calcite generally is attributed to the formation of caliche in the interbeds (Rightmire and Lewis, 1987b). Rightmire and Lewis (1987b, p. 35-36) reported trace amounts of iron oxyhydroxides, hematite, siderite, and dolomite in some samples. C.T. Rightmire and B.D. Lewis (USGS, written commun., 1995) reported one tentative identification of the zeolite mineral chabazite. This lack of zeolite in the system probably precludes zeolite exchange with radionuclides in solution as a major factor in preventing the migration of radionuclides to the aquifer.

Analyses of the clay minerals indicate that illite is predominate (ranging from 10-100 percent of the clay minerals identified), and that lesser amounts of smectite, mixed-layer illite/smectite, kaolinite, and possibly chlorite are present. Illite generally is considered detrital; but fluctuations in the amounts of smectite and mixed-layer clays, the presence of altered potassium feldspars, and the possibility of thermal alteration, suggest that some illite and also smectite is forming (Rightmire and Lewis,



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1987b). The water chemistry also is favorable for the precipitation of these minerals. The clay minerals that are present in the system provide sites for sorption and/or ion exchange with any water containing radionuclides moving in the system. These processes would prevent migration of radionuclides to the aquifer. Conditions that need to be present to prevent sorption or exchange of radionuclides with clay minerals will be discussed in more detail in chapters 4 and 5.

### 23.4.4 Pedogenesis

Weakly-developed to well-developed paleosols have been identified in the surficial sediment and sedimentary interbed A-B and, although not documented, probably also occur in interbeds B-C and C-D. Forman and others (1993) identified paleosols associated with young loess units in two excavations at the SDA; one of these paleosols is thought to represent a prolonged period of pedogenesis, possibly exceeding 20 thousand years. These units contain A, B, and C soil horizons, carbonized zones, and abundant clay cutans having a maximum clay concentration of from 24 to 36 percent. Carbonized zones represent vegetation that was inundated and baked by lava flows during the geologic past. The distribution and characteristics of these zones have not been described on the basis of sediment cores from the SDA. Carbonate content of these paleosols generally is smaller than 10 percent but is as large as about 20 to 25 percent in some zones. Mineralogical analyses of the sediments indicate that most of the carbonate in the system is in the form of calcite, but some dolomite also was identified (Bartholomay and others, 1989). Mineralogical analyses of core samples of the surficial sediment and sedimentary interbeds A-B, B-C, and C-D reported by Barraclough and others (1976) show carbonate in the form of calcite. Calcite contents range from absent for most of the samples to 54 percent for others. Calcite content larger than 10 percent was reported for interbed C-D in wells USGS 88, 89, 90, and 96; for surficial sediment in wells USGS 92 and 95; and from a vesicle at about 9 m in well USGS 9 (figs. 2-2 and 2-3). Subsequent analyses by Rightmire and Lewis (1987b) showed carbonate contents as large as about 12 percent in interbed B-C, as large as about 29 percent in interbed C-D in well USGS 93A, and as large as about 2 percent in interbeds B-C and C-D in well USGS 96B (Rightmire, 1984). The largest carbonate content in interbed B-C in well USGS 93A is at the base of the interbed.

### 23.4.5 Thickness and areal extent

Elevations of the tops and thicknesses of the surficial sediment and at least 10 sedimentary interbeds in 94 wells

at and near the SDA were described by Anderson and Lewis (1989), Anderson and others (1996), and Magnuson and Sondrup (1998). Of these sedimentary units, only the surficial sediment and sedimentary interbeds A-B, B-C, C-D, and F-G are substantially thick and areally extensive. Surficial sediment is present in all 94 wells, has a top elevation that ranges from 1,514 to 1,544 m above sea level, and is up to 8 m thick. Sedimentary interbed A-B is present in 44 wells, has a top elevation that ranges from 1,511 to 1,528 m above sea level, and ranges in thickness from 0 to 6 m. Sedimentary interbed B-C is present in 91 wells, has a top elevation that ranges from 1,474 to 1,504 m above sea level, and ranges in thickness from 0 to 10 m. Interbed C-D is present in 76 wells, has a top elevation that ranges from 1,443 to 1,469 m above sea level, and ranges in thickness from 0 to 11 m. Sedimentary interbed F-G is near the base of the unsaturated zone and is about 90 m deeper than interbed C-D. Interbed F-G is present in 12 of 15 deep wells near the SDA, has a top elevation that ranges from 1,347 to 1,373 m above sea level, and ranges in thickness from 0 to 6 m. The surficial sediment and all sedimentary interbeds contain known gaps that are related to the irregular surfaces of underlying basalt flows. Interbeds B-C and C-D have the fewest known gaps and dip gently southeastward across the SDA. Surface reliefs of interbeds B-C and C-D, between well NA-89-1, near spreading area A located approximately 1.5 km west of the SDA (fig. 1-1), and well USGS 90, near the eastern side of the SDA, are 8 and 18 m, respectively. Perched ground water in wells at the SDA is present above and within interbeds A-B, B-C, and C-D (Rightmire and Lewis, 1987b; Magnuson and Sondrup, 1998) and may be related partly to the lateral movement of infiltrating water from the spreading areas to the SDA (Rightmire and Lewis, 1987b).

### 23.4.6 Hydraulic conductivity

Hydraulic conductivity, an indirect measure of sediment lithology, varies more than four orders of magnitude in the surficial sediment, more than three orders of magnitude in interbed A-B, more than six orders of magnitude in interbed B-C, and more than seven orders of magnitude in interbed C-D (Barraclough and others, 1976; McCarthy and McElroy, 1995). Barraclough and others (1976, p. 50) reported a range of vertical hydraulic conductivity for these sedimentary units in 10 wells, USGS 87, 88, 89, 90, 91, 92, 93, 94, 95, and 96 (figs. 2-2 and 2-3), of from  $1.6 \times 10^{-7}$  to 3.0 m/d. The smallest vertical hydraulic conductivity reported for interbed C-D in each of these wells ranges from  $8.0 \times 10^{-7}$  m/d in well USGS 88 to  $2.6 \times 10^{-1}$  m/d in well USGS 91. The largest

vertical hydraulic conductivity reported for interbed C-D in each of these wells ranges from  $6.1 \times 10^{-5}$  m/d in well USGS 89 to  $9.3 \times 10^{-1}$  m/d in well USGS 95. The largest range of vertical hydraulic conductivity reported for interbed C-D was for well USGS 88,  $8.0 \times 10^{-7}$  to  $1.3 \times 10^{-1}$  m/d (Barracough and others, 1976). These ranges of hydraulic conductivity probably reflect centimeter-scale changes in lithology, from clay- to gravel-sized clasts, within the surficial sediment and sedimentary interbeds A-B, B-C, and C-D.

### 2.3.5 Hydrologic pathways

Sedimentary interbeds at the SDA are characterized by abrupt changes in thickness related to the topography of underlying basalt flows (Anderson and Lewis, 1989). Most of these flows are pahoehoe flows, which commonly contain inflation and deflation features, including ridges and closed depressions, and vary in relief a few meters within distances of a few tens of meters. Anderson and Lewis (1989, p. 38 and 47) described one such structure on the top of basalt-flow group C in the western part of the SDA, where the thickness of sedimentary interbed B-C changes abruptly from zero on a basalt ridge to more than 6 m in an adjacent depression. Rightmire and Lewis (1987a) suggested, on the basis of carbonate encapsulating clay pellets covered with desiccation cracks present in interbed B-C, that sediment and water collected in a topographic depression at the present-day location of wells 76-4 and 76-4A during the deposition of interbed B-C. Infiltrating water may migrate towards such depressions at some sediment-basalt interfaces, possibly resulting in areas of localized perched water and/or preferential flow. Some basalt ridges, such as those on the top of basalt-flow group C in the western part of the SDA, that are penetrated by wells 76-1, 76-2, and 78-3 (Anderson and Lewis, 1989, p. 38 and 47), also may be areas of preferential flow. This is because these ridges were not covered by sediment and probably are inflated lobes of pahoehoe basalt cut by large cooling fractures.

Vertical water movement through the basalt-sedimentary sequence is largely controlled by the hydraulic conductivity of fractures in the basalt flows and by the smaller hydraulic conductivity of some of the finer sediments in the interbeds. An additional and even greater constraint occurs at the base of each sedimentary layer, at the sediment-basalt interface. "This is caused by the discontinuity of pore spaces from the sediment to the basalt, due both to the lower porosity of the basalt and to the relatively great distances between its fractures. In other words, at the interface, perhaps only 10 percent of the

basalt surface is composed of permeable openings, and these are partially filled by sediment. The other 90 percent is virtually impermeable. This, in effect, provides a thin skin that is estimated to have one-tenth or less of the permeability of the sediments alone" (Barracough and others, 1976).

Horizontal water movement through the basalt-sediment sequence is largely controlled by the large hydraulic conductivity associated with basalt fractures, rubble, and scoria, a term synonymous with cinders. In the saturated zone, these features provide the main conduits for ground-water flow. In the unsaturated zone, fractures, rubble, and scoria have the potential to channel perched water over large distances within short periods of time. Estimates of ratios of horizontal to vertical basalt permeabilities at the INEEL range from about 3:1 to 300:1 (Barracough and others, 1976; Magnuson, 1995; Magnuson and Sondrup, 1998).

The presence, absence, or transient nature of perched water in wells at the SDA may be the result of several geologic factors. Cecil and others (1991) suggested four possible mechanisms to explain zones of perched water. These include: (1) contrasts in vertical hydraulic conductivity between basalt flows and sedimentary interbeds, (2) decreased hydraulic conductivity in interflow baked zones, (3) decreased vertical hydraulic conductivity in dense, unfractured basalt, and (4) decreased vertical hydraulic conductivity from sedimentary and chemical filling of fractures in basalt. Cecil and others (1991, p. 23) demonstrated that perching can take place within sedimentary interbeds and basalt flows.

The recurring presence of a ground-water mound beneath the spreading areas indicates that most of the water from this source moves vertically to the water table (Rightmire and Lewis, 1987b). However, the lateral continuity of some of the interbeds, particularly the C-D interbed, suggests that water recharged from the spreading areas also could move to the east the approximately 1.5 km necessary for it to be present in the unsaturated zone beneath the western edge of the SDA. Rightmire and Lewis (1987b) reported the presence of water coinciding with the A-B, B-C, and C-D interbeds on the northwest side of the SDA, and it is conceivable that this water moved laterally from the spreading areas in a stepwise depth progression from the shallowest to deepest interbeds. If so, there is possibly significant lateral movement of water from beneath the spreading areas. Thus, it is possible that the perched water beneath the SDA is derived largely from the spreading areas and (or) the BLR and receives only a minor contribution of moisture from ver-

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tical water movement derived from atmospheric precipitation.

Basalt fractures, rubble, and scoria are present in the subsurface at the SDA, but their locations and orientations are poorly known. Simple cooling fractures likely are present in all basalt flows, but large fractures, such as those along the edges of inflated pahoehoe flows, probably are very localized. These fractures, which can be as much as a few meters wide and tens of meters long, are oriented in the direction that each lobe of pahoehoe lava traveled during its emplacement, inflation, and cooling. These directions are only approximately known for basalt-flow groups A and B. Basalt rubble in some wells has been described but has not been systematically characterized. Magnuson and Sondrup (1998) described a rubble zone across the SDA at a depth of about 60 m that could affect the distribution of carbon tetrachloride. The potential for scoria, a near-vent deposit, is greatest in basalt-flow group C. This is because its vent is thought to be in the subsurface near the SDA (Anderson and Liszewski, 1997, p. 10). Rightmire and Lewis (1987a, p. 64) reported the presence of grout cement in well 76-3, which likely was carried by water through a cinder (scoria) zone beneath interbed B-C from well USGS 93 to well 76-3, a distance of 168 m. Basalt fractures, rubble, and scoria probably affect the potential for perched water in complex ways.

## 2.4 Hydrologic framework

### 2.4.1 Meteorology

The INEEL receives an average of about 22 cm of precipitation each year. On average, May and June are the wettest months and July is the driest month (Robertson and others, 1974, p. 8). Normal annual snowfall is about 66 cm and accounts for about 30 percent of the annual precipitation.

### 2.4.2 Surface water and local runoff

#### 2.4.2.1 Areal infiltration

Areal recharge on the eastern SRP from rainfall and snowmelt infiltration generally is considered to be small. Cecil and others (1992, p. 709) calculated net areal infiltration rates at the SDA to range from 0.36 to 1.10 cm/yr, or 2 to 5 percent of the long-term annual precipitation. However, topographic features locally may focus areal recharge in lower areas. The SDA is located in a topographic depression. Barraclough and others (1976, p. 8)

reported that the SDA has been flooded at least twice (1962 and 1969) by local runoff from rapid spring thaws. Heavy rainfall and melting snow within the SDA have also introduced water into trenches and pits.

Areal recharge may be affected by changes in soil properties introduced by construction of waste trenches in surficial sediments at the SDA. Shakofsky (1995, p. 24) determined that the hydraulic conductivity of disturbed soils overlying trenches in the vicinity of the SDA and corresponding recharge from areal precipitation, generally are larger than the hydraulic conductivity of and recharge through nearby undisturbed soils.

#### 2.4.2.2 Stream infiltration

It has long been recognized that the BLR is an important component of the hydrologic system that controls the fate and transport of waste constituents in the subsurface at the INEEL. Robertson (1974, p. 26) noted that recharge from the BLR is the "biggest hydrologic variable" in determining the future behavior of waste constituents in the aquifer.

Since 1965, a large proportion of flow in the BLR has been diverted to the INEEL spreading areas for flood control. During 1982–85, approximately two-thirds of the flow that entered the INEEL was diverted to the spreading areas (Pittman and others, 1988, p. 18). Rapid infiltration of these diverted flows locally affects water levels in the SRP aquifer. Pittman and others (1988, p. 18) observed that the water table in the vicinity of the SDA rose as much as 4.9 m during 1982–85 in response to recharge from surface water diverted to the spreading areas.

Some evidence exists that rapid infiltration also may produce significant lateral flow in the unsaturated zone. Barraclough and others (1976, p. 51) noted the presence of perched water in several boreholes within the SDA. They observed that an extensive zone of saturated to nearly saturated basalt existed beneath the burial ground. The source of water was not known, but they believed that thin perched-water zones could represent long-term local accumulation of percolating precipitation or more rapid recharge from the 1969 flood at the SDA. They suggested that isotopic analyses might reveal the source and history of the perched water.

Rightmire and Lewis (1987, p. 83) noted that an anomalously light isotopic content in water samples from perched water samples at the SDA may be attributed to a water source at an altitude higher than the surface of the SRP. They stated (1987, p. 1) that "...stable isotope and chemical data suggest that the perched water observed beneath the SDA is not due to vertical infiltration through the ground surface at the SDA, but is due to lateral flow of

water that infiltrated through the diversion ponds." They hypothesized at that time that water accumulates as a perched mound on a thick, continuous sedimentary interbed at a depth of 73 m and then moves laterally to the SDA 1.5 km to the northeast.

Rightmire (1984, p. 32) observed that "...sedimentary lining and filling of fractures is the result of water-borne sedimentation. Layers of oriented clay particles overlain by disoriented coarser material suggest a series of minor recharge events followed by a major recharge event to fill the fractures." Rightmire also noted that the presence of cement grout in fractures penetrated in a test hole demonstrates that open fractures occur at depths of at least 54 m. These fractures would permit transport of sediments and fluids through the unsaturated zone.

Anderson and Lewis (1989, p. 20) noted that basalt flows contain numerous fractures that may provide unrestricted avenues for vertical and horizontal flow of contaminated water. They suggested that sediment layers may control vertical flow depending on grain size and sorting characteristics. They noted that lateral flow and perching of water may take place along some clay and silt layers, and that discontinuous layers may divert flow toward underlying or adjacent basalt flows. They also stated that the potential for lateral flow away from the SDA along the east-sloping surface of the C-D interbed is large. Anderson and Lewis stated that additional evaluation of rock and sediment characteristics, including the distribution and characterization of flow contacts, fractures, and vesicles, and the lithology of major sedimentary interbeds, is needed to determine the potential for contaminant migration to the Snake River Plain aquifer.

### 2.4.3 Unsaturated-zone flow

The unsaturated zone is defined as the portion of the subsurface between the land surface and the lowest water table. The 200 m thick unsaturated zone at the SDA includes both saturated conditions (where perching occurs; discussed in section 2.4.3.2) and unsaturated conditions.

#### 2.4.3.1 Flow in a homogeneous medium

For water driven by gravity and pressure gradients when the medium is saturated, as from ponded infiltration or perching, Darcy's law can quantify the flow as:

$$q = -K_{sat} [\nabla \psi + \rho g] \quad (2.1)$$

where:

$q$  = flux density, in volume rate of flow per unit area;

$K_{sat}$  = saturated hydraulic conductivity, in length per time;

$\psi$  = water pressure in length;

$\rho$  = density of water, in mass per volume; and

$g$  = gravitational acceleration, in length per time squared.

For simplicity, this equation frequently is applied with  $\rho g$ , the weight of water per unit volume, set equal to dimensionless unity. Then  $\psi$  has dimensions of length (m), and  $K$  of length per time (m/s).

If the medium is unsaturated and the flow is diffuse, an unsaturated form of Darcy's law can quantify the flow at a point in space and time.

In one-dimension:

$$q = -K(\theta) \left[ \frac{d\psi}{dz} + \rho g \right] \quad (2.2)$$

where:

$\theta$  = water content, in volume of water per total volume;

$z$  = vertical distance, in length;

$K$  = hydraulic conductivity, in length per time.

$K$  depends strongly and nonlinearly on  $\theta$ . The matric pressure ( $\psi$ ) is of particular interest because it often determines the chief transport processes in an unsaturated medium. It arises from the interaction of water with a rigid matrix. Matric pressure, defined very precisely by Bolt and others (1976), may be thought of as the pressure of the water in a pore of the medium relative to the pressure of the air. When a medium is unsaturated, the water generally is at lower pressure than the air, so the matric pressure is negative. Darcy's law applies directly in field situations where flow is steady, though steadiness in actuality is often temporary or approximate. In steady conditions equation 2.2 with a single  $K(\theta)$  function can describe the flow within any homogeneous portion of the medium.

When unsaturated flow is transient (nonsteady), as it generally is, the flow itself causes the water content to change throughout the medium, which leads to continuously changing hydraulic conductivity and driving forces. These effects can be accommodated mathematically by combining the equation of continuity with Darcy's law (2.2) to get Richards' (1931) equation, which for one-dimensional vertical flow can be written:

$$C(\psi) \frac{\partial \psi}{\partial t} = \frac{\partial}{\partial z} \left( K \frac{\partial \psi}{\partial z} \right) + \rho g \frac{\partial K}{\partial z} \quad (2.3)$$

where:

$C$  = differential water capacity, a property of the medium defined as  $d\theta/d\psi$ .

With certain implicit assumptions, such as homogeneity of the medium, Richards' equation keeps track of the whole dynamic, unsteady flow process. In practice, most mathematical models of unsaturated flow assume equa-

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tion 2.3 applies, whether appropriately or not. The equation is nonlinear and can be solved numerically in the general case (Lappala and others, 1987; Healy, 1990) or analytically (Salvucci, 1996) with simplifying assumptions that may be suitable in some, but perhaps not most, applications.

### 24.3.2 Flow and perching in stratified unsaturated zones

Contacts between layers that contrast in hydraulic properties commonly impede vertical flow. Such impediments occur most obviously when water moves down from a coarse to a fine layer, as from coarse sand to silt. If both layers are near saturation, the fine layer has smaller hydraulic conductivity; therefore, flow slows when it reaches the fine layer. If the coarse layer is nearly saturated but the fine layer is initially fairly dry, it is possible for flow to be initially dominated by the sorptive nature of the fine medium, which tends to suck water out of the coarse material. In the latter case flow is not impeded by the fine layer until more uniformly saturated conditions occur.

Where a fine layer overlies a coarse layer, water moving downward is impeded under many conditions. When coarse material is dry, it has an extremely small hydraulic conductivity; thus it tends not to admit water into the pores and exhibits a somewhat self-perpetuating resistance to flow. Water breaks into the coarse layer if the pressure at the layer interface builds to the point that the water-entry pressure (the minimum water pressure needed to fill an empty pore) of some of the large pores is exceeded. This can generate instabilities, as discussed in section 2.4.3.3. Stable or not, water flowing into the pores of the coarse medium increases its hydraulic conductivity drastically. Then the wetter coarse medium will conduct more easily, and will progress toward steady-state flow. The effective hydraulic conductivities of the two layers will then be essentially equal. In the approach to steady flow, the water content in each layer adjusts to accommodate this equalizing of conductivities. The water content of the coarse medium, however, does not increase enough to make the effective hydraulic conductivity as large as it would be for a single-layered system composed of fine material only. Thus, typical flow through layers where fine overlies coarse is slower than it would be if both layers had the properties of the fine medium. Miller and Gardner (1962) demonstrated this effect experimentally.

A thin layer of small hydraulic conductivity can limit downward flow to the point of being the dominant influence on flow through the sequence. Stothoff (1997) considered a granular medium above a fractured bedrock (similar to much of the INEEL unsaturated zone). The

bedrock admits water only under nearly saturated conditions. Stothoff's interpretation assumes the fractures are of greater-than-microscopic width and the rock is otherwise impermeable. The thickness of the granular layer strongly influences the fraction of average precipitation that flows into the bedrock (and presumably further, to the aquifer); thus, a thin alluvial layer more easily becomes saturated to the layer interface and hence more frequently permits deep percolation.

Perching, an accumulation of water in a region of the unsaturated zone such that it becomes locally saturated even though there is unsaturated material below, is not unusual in the unsaturated zone at the INEEL. The artificial infiltration of wastewater has created perched zones that have persisted for several years (Orr, USGS written commun., 1999). Perching usually results from a large flux of water that encounters a severely impeding layer. It may be a temporary or permanent feature, depending on the nature of the medium, the prevailing hydrologic conditions, and the effect of artificial modifications. Perching complicates a contamination problem in several ways. The high water content of a perched zone causes greater hydraulic conductivity and potentially faster transport through the three-dimensional system. The main effect is not a direct increase in vertical flux because the increase in effective vertical hydraulic conductivity is offset by a diminished vertical hydraulic gradient within the perched water. (Vertical flux within and below the perched water cannot be faster than the vertical flux above the perched water or the perched water would have drained). Horizontally, however, there may be greatly increased flow. This enhanced horizontal flow may in turn bring much water to points where the vertical conductivity is locally large (for example, where there is a vertical fracture), or to where perching occurs at a sloping interface along which the enhanced flow has a substantial vertical component. Perched zones commonly are large in horizontal extent, so these effects related to horizontal flow can become more important than vertical flow for contaminant transport. New and different processes may significantly affect contaminant transport in a perched zone. Reduced aeration, for example, may affect biochemical processes. At the scale of the entire stratified unsaturated zone, perching may significantly increase anisotropy.

### 24.3.3 Preferential flow

In an unsaturated medium, flowpaths that permit faster transport than other paths are common. Flow through these preferential paths is called preferential flow. Such a path may be a single pore, a connected series of pores, or a group of adjacent pores acting in parallel. A

path may be preferential because of either the permanent character (for instance pore size) or the present state (for instance water content) of its pores. The path may be a single pore, a connected series of pores, or a group of adjacent pores acting in parallel.

Preferential flow, although difficult to observe or sample, can convectively transport contaminants to the aquifer far sooner than might be predicted on the basis of ordinary analysis of bulk medium properties and Richards' equation (2.3). Additionally, if the flow is entirely or primarily through preferential paths, the interaction of contaminants with solid material is effectively limited to a fraction of the subsurface medium. This limits adsorption and other attenuating processes. Thus, preferential flow decreases both the time and the space available for contaminant attenuation, chemical or radioactive decay.

Various mechanisms cause preferential flow (Steenhuis and others, 1995; Or, 1996). Three kinds of preferential flow and the contributing mechanisms are: (1) macropore (or short-circuiting) flow, caused by flow-enhancing features of the medium, (2) funneled (or deflected or focused) flow, caused by flow-impeding features of the medium, and (3) unstable (or fingered) flow, caused by flow-enhancing conditions of parts of the medium at the time of interest. This categorization is similar to several others in widespread use and includes, directly or indirectly, essentially every known process that might be considered preferential-flow phenomenon. As for other subjects for which theory is in the early stages of development, the terminology classifying preferential flow often is imprecise or ambiguous. For example, focused flow might refer to funneled flow or to flow in certain types of macropores. Sometimes distinctions between flow types blur. Often several mechanisms are acting within the same medium.

#### 2.4.3.3.1 Macropore flow—heterogeneity of the porous medium

Macropores, which are distinguishable from other pores by their larger size, greater continuity, or other attributes, conduct preferential flow under some conditions (Luxmoore, 1981), such as extreme wetness. Common macropores include wormholes, rootholes, and fractures. These may be visible by eye, or difficult to detect without special techniques. Where macropore flow occurs, flow through the remainder of the medium may be called matrix flow. Macropore flow may be interpreted to include preferential flow arising not only from individual large pores but also from the areal heterogeneity of bulk properties. Hydraulic conductivity, wettability, and other flow-affecting properties generally vary from place to

place, so that flow rates are greater in some locations than others.

The speed and behavior of macropore flow vary. When macropores are filled, flow is generally fast. When macropores are empty, they constitute a barrier to matrix flow and there may be essentially no flow through the macropores themselves (Montazer and Wilson, 1984). In some conditions, however, there may be significant film flow along fracture walls (Tokunaga and Wan, 1997). In macropores that are partly filled, there are many possibilities for the configuration and flow behavior of water (Su and others, 1999; Nicholl and others, 1994; Pruess, 1998; Nitao and Buschek, 1991; Persoff and Pruess, 1995). These possibilities and their effect on water and contaminant transport are not yet well understood. Recent research of Faybishenko (1999) suggests that there may be chaotic fluctuations in this type of flow, which in turn suggests the possibility of new mathematical treatments.

#### 2.4.3.3.2 Funneled flow—heterogeneity of the porous medium

Layers or lenses of material that, to some degree, impede vertical flow because of textural contrasts or other reasons can cause horizontal flow. Often such layers are tilted so that gravity causes flow to move in a particular lateral direction. Whether or not tilting is a factor, both the lateral flow and the vertical flow that moves through or around the impeding feature normally follow preferential paths.

Several studies have investigated the effects of these impeding features in generating preferential flow. Some of the first rigorous research was by Kung (1990a, 1990b). In field experiments at a site with sandy soil, Kung found that the flow became more preferential with depth. At about 6-m depth, the flow was moving through less than 1 percent of the whole soil matrix. Although this medium had no significant observable macropores, preferential flowpaths were shown to be the dominant flow pattern. The main feature causing this preferential flow was "...an interbedded soil structure with textural discontinuities and inclined bedding planes." Striegl and others (1996) hypothesized that funneled flow could have transported tritium and carbon-14 contaminants over large horizontal distances in the unsaturated zone at a waste-disposal site near Beatty, Nevada. Considering the work of Kung and other researchers, Pruess (1998) noted that because funneling can produce very rapid flow and results from horizontal impediments "...we have the remarkable situation that unsaturated seepage can actually proceed faster in a medium with lower average permeability."

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### 2.4.3.3.3 Unstable flow—heterogeneities in the condition of the medium

An unstable variation in flow and water content can increase hydraulic conductivity in some parts of the medium even if the fundamental hydraulic properties of the medium are uniform. Usually, texturally contrasting layers contribute to this unstable flow. The classic case is a layer of fine material above coarse material. In this case infiltrating water does not immediately cross the interface into the coarse material. When water pressure builds up significantly at the interface, water may break through into the coarse medium at only a few points, as discussed in section 2.4.3.2. The material near individual points of breakthrough becomes wetter and hence much more conductive. For some time thereafter, additional flow into the coarse material moves in the few “fingers” that are already wet. Between fingers, the medium remains relatively dry for some time. In addition to textural contrasts, hydrophobicity or water repellency of the medium may contribute to unstable flow (Dekker and Ritsema, 1994; Ritsema and Dekker, 1994; de Rooij, 1995; Ritsema and others, 1998). Unstable flow may also be associated with air trapping (Wang and others, 1998a). Several researchers (Glass and others, 1988; Baker and Hillel, 1990; Yao and Hendrickx, 1996) have explored the general types of behavior expected in unstable flow.

Preferential flowpaths may be interrupted by layer boundaries. Numerical experiments by Russo and others (1998) suggest that the spatial variability of soil properties and the temporal variability of infiltration and redistribution both may act to homogenize transport phenomena. Shakofsky and Nimmo (1996) found that flow through undisturbed surficial soil at the INEEL appeared to be fast and highly preferential near the land surface, but was drastically slowed at about a 2-m depth, apparently because of a layer at this depth that differed in hydraulic properties. In nearby soil that had been disturbed and homogenized much like the soil at the SDA landfill, the flow was less preferential and there was no evidence of sudden slowing at a particular depth. It is possible for processes to be operative that homogenize preferential flow. How common these processes are, however, is currently a matter of speculation among unsaturated-zone hydrologists.

### 2.4.3.4 Quantitative treatment of preferential flow

#### 2.4.3.4.1 Effective properties

A widely used approach to mathematical modeling when some or all of the flow is preferential is to assume that the effective hydraulic properties of a large volume of the medium are equivalent to the average properties of a

homogeneous granular porous medium. The effective hydraulic properties then can be applied directly in numerical simulators developed for diffuse flow in granular media based on Darcy’s law and Richards’ equation. This technique is often called the “equivalent-porous-medium” approach. In this report it is called the “equivalent-granular-medium” approach to prevent confusion with the fact that the basalts at the INEEL intrinsically are porous media themselves.

The equivalent-granular-medium approach is valid only if certain conditions are met. For example, the portion of the unsaturated zone under consideration must be divisible into finite volumes within each of which the net flow, including all preferential flow, is representative of the medium as a whole. Volumes taken too small will invalidate the approach because they have different amounts of preferential flow within them and therefore are not representative of the whole medium. On the other hand, if these finite volumes are so large that only a small fraction of the assumed volume is participating in the flow, the actual transport velocities can be much greater than the modeled velocities. In practice, an adjustment of the “effective porosity” of the equivalent granular medium is commonly done to compensate for such effects, though this adjustment will not produce accurate predictions if, as expected, the degree of preferential flow depends strongly on the degree of saturation. Bear (1978, p. 28–31) gives a detailed explanation of volume and homogeneity considerations for continuum approaches in general. Pruess (1998) describes in detail some of the possible large-scale transport-accelerating mechanisms associated with preferential flow. In many applications, the equivalent-granular-medium approach is the only one that is practical and has a significant history of prior use. Other approaches may be less subject to fundamental limitations but are not sufficiently developed or tested to make predictions with a high level of confidence.

The main advantage of the equivalent-granular-medium approach is that many existing theories, models, and familiar techniques can be immediately applied. A recent example is that of Reitsma and Kueper (1994), who found that the Brooks-Corey model (Brooks and Corey, 1996) of soil water retention, which was formulated to apply to ordinary soil materials, well represented measured retention properties of a rough-walled rock fracture. The equivalent-granular-medium approach has additional drawbacks, mostly because the flow theory and models involved were developed for wet, homogeneous granular, porous media. Preferential flow may be too irregular to be described accurately by a model in which averaged values of hydraulic properties have been applied. For example



Ma and Selim (1996) noted that averaging the flow rate and using the average to quantify solute transport is not an adequate method; using the actual distribution of flow rates is more suitable. The equivalent-granular-medium approach also is inadequate wherever it is essential to have knowledge of the specific flowpaths that may act as conduits for most of the contaminant transport.

#### 2.4.3.4.2 Dual-modality and multi-modality

A family of approaches that rely on a conceptual partitioning of water or pore space into portions with different flow rates and behaviors and given names such as "dual-porosity" and "dual-modality", may represent more realistically flow that includes preferential paths than do traditional unsaturated-flow models (Reedy and others, 1996). Magnuson and Sondrup (1998, p. 4-48) used a model of this type in calibrating for volatile organic contaminants (VOCs). The concept of mobility, that is, how easily moved certain system components (in particular, contaminants) are in different parts of the medium, is frequently used in defining and characterizing these approaches.

Some of these models are used more commonly for solute transport than for water flow in preferential paths. The simplest ones assume matrix flow to be negligible, so that all flow is preferential flow. Given the nonlinear nature of unsaturated flow, the difference in conductance between, say, a wormhole and an intraaggregate pore between clay particles may be several orders of magnitude and, for practical purposes, may constitute a mobile-immobile distinction (van Genuchten and Wierenga, 1976). Other models assume the matrix to be permeable but with different properties and possibly different modes of flow than the portion of the medium that has preferential flow. In extensive field experiments, Ghodrati and Jury (1992, fig. 2) showed effects of preferential contaminant transport and noted that existing transport models for nonhomogeneous soils were inapplicable. They concluded that "...new approaches will have to be developed to address preferential flow when the soil matrix is also permeable enough to permit transport of water and chemicals through it."

The degrees of possible mobility cover a continuum, and truly immobile water is unlikely. Some models postulate as many as three degrees of effective mobility; a few models postulate more. For example mesopores in addition to micropores and macropores, may be considered (Luxmoore, 1981). A closer approach to a continuum of mobility is that of Griffioen and others (1998), who worked toward a parameterization of mobility.

Other recent models were described by Jaynes and others (1995), and Casey and others (1997). It is also possible to combine mobile-immobile models with other models, especially for reactive solute transport (Ma and Selim, 1997).

#### 2.4.3.4.3 Additional considerations in the quantification of unstable flow

Quantification of unstable flow is complicated by two factors that do not apply to macropore or funneled flow: (1) unstable flow is not tied to particular permanent features of the medium, and (2) the preferentiality of unstable flow changes dynamically (for instance, flowpaths commonly would grow wider as flow progresses through them). Theories of unstable flow in terms of scaling (Miller and Miller, 1956) and other concepts, have been developed in studies by Glass and others (1989a, 1989b), Raats (1973), Philip and Forrester (1975), Parlange and Hillel (1976), Diment and others (1982), Hillel and Baker (1988), and Selker and others (1992a, 1992b).

#### 2.4.3.5 Contentions and ambiguities concerning preferential flow

At present (1999) there is disagreement in the scientific community concerning the prevalence of preferential flow. Statements in many recent publications suggest that preferential flow is uncommon: "Pulses of infiltration are... naturally damped in a diffusion-like process" (Stephens, 1996, p. 97); "Field studies that demonstrate preferential flow are restricted mostly to fractured rock and root zones in arid regions" (Scanlon and others, 1997); "Fractures are unlikely to provide rapid, open channels for flow" (Soll and Birdsell, 1998); "It seems likely that the strong lateral potential gradients that result from preferential flow into the dry subsoil rapidly damp out irregular wetting fronts" (Phillips, 1994); "Soil hydraulic properties, initial soil water content, and infiltration rate exert an important control on instability" (Hendrickx and Yao, 1998).

Other statements argue for the prevalence of preferential flow: "The occurrence of preferential flow is the rule rather than the exception" (Flury and others, 1994); "Preferential flow is a dominant feature of structured soils, particularly those with pronounced layering" (Jury and Wang, 1999); "A most effective condition for fast preferential flow is the presence of sub-horizontal barriers of significant length" (Pruess, 1998); "It is now apparent that fingered flow should be expected in any sandy soil with unsaturated flow" (Selker and others, 1991); "While once believed to occur exclusively in structured soils, preferential flow is now recognized as prevalent under a wide range of conditions in permeable, structureless (for



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example, devoid of macropore structure) soils” (Jury and Wang, 1999).

In virtually every unsaturated-zone transport problem, it is essential to assess the prevalence of preferential flow and the flow mechanisms that might be active. One approach is to evaluate the features of a particular site that might cause macropore, funneled, or unstable flow. For example, for unstable flow, Wang and others (1998a, 1998b) analyzed the likelihood of a variety of mechanisms that might cause unstable flow. Another approach is to collect and evaluate evidence from observed water or solute distributions that cannot easily be explained without hypothesizing preferential flow.

### 2.4.3.6 Flow in the unsaturated zone at and near the Subsurface Disposal Area

#### 2.4.3.6.1 Surficial sediments

The unsaturated hydraulic conductivity and water retention of the surficial sediments in the unsaturated zone at the INEEL are fairly typical of a medium in which structural features such as aggregation and macropores have evolved over time (Shakofsky, 1995; Nimmo and others, 1999). At the INEEL, much of the surficial sediment is in the silt loam textural class. At many locations, for example at the SDA, the hydraulic properties have been substantially altered by mechanical disturbances. The main known effects of these disturbances are less macropore flow and less stratification. Like other surface soils with developed structural features, the hydraulic conductivity of these sediments, disturbed or not, may exhibit substantial local anisotropy.

Available evidence suggests that flow in the unsaturated zone is highly preferential where the soil is undisturbed and preferential to some degree elsewhere. Especially under wet conditions during or shortly after infiltration, the flow may be dominated by macropores. Natural layers within these sediments, where they are undisturbed, and the interface with the basalt in all locations may significantly slow downward flow. This retarding influence may be especially important at shallow depths, where its effect is to retain more water high in the root zone where it is more likely to be removed by evapotranspiration.

#### 2.4.3.6.2 Basalts

The hydraulic properties of the fractured basalts have been represented by the effective properties approach (Magnuson and Sondrup, 1998). The effective hydraulic conductivity has not been determined by direct measurements but rather through inverse modeling, as discussed

in chapter 6. There is likely to be substantial anisotropy in these basalts, large and small, though it is difficult to estimate its nature and degree.

Flow within the basalts is dominated by macropore flow through fractures. The basalt matrix is sometimes assumed impermeable, which for many purposes may be an adequate approximation. A large body of evidence, including that of the Large-Scale Infiltration Test (LSIT) (Dunnivant and others, 1998), indicates that when the basalt fractures are filled, they can conduct rapidly, perhaps meters per day or faster. Some fractures become filled from wetting events that might occur as often as several times per year. There is also evidence that some fractures, especially ones with dead ends, do not conduct significant flow, or at least do not allow significant downward flow (Dunnivant and others, 1998). Little is known about the processes or rates of flow in parts of basalt in which all fractures are unsaturated. A common assumption, possibly valid for some purposes, is that such flow is negligible compared with episodic flow through filled macropores.

#### 2.4.3.6.3 Sedimentary interbeds

The sedimentary interbeds closely resemble the surficial sediments in some ways, but they differ significantly, for example in having greater density and more uniform structure. They probably have no significant wormholes or rootholes and may be less aggregated than the surficial sediments. The sedimentary interbeds are highly stratified; internal layers and lenses differ substantially from each other in texture and structure in terms of such features as baked zones. The hydraulic properties of interbeds would tend to be like those of the surficial sediments except for systematic differences such as smaller hydraulic conductivity resulting from greater bulk density. Another difference from surficial sediments is that interbed sediments are likely to retain more water after episodes of drainage. Because the sediments are granular, the interbeds may be less anisotropic within internal layers than other parts of the unsaturated zone. At scales of a few meters or greater, basalt-interbed interfaces lead to great effective anisotropy.

Flow behavior in sedimentary interbeds probably varies widely both seasonally and sporadically as water content and hydraulic conductivity vary. At least to some degree, sedimentary interbeds are likely to impede vertical flow and to cause preferential flow from basalt fractures to become more diffuse. The interbeds probably have less macropore flow within them than the surficial sediments or basalts, but their layered structure may be conducive to funneled or unstable flow. Thus it is likely

that, to some degree, they cause vertical and horizontal preferential flow.

Saturated hydraulic conductivity, an indirect measure of sediment lithology, ranges more than four orders of magnitude in the surficial sediment, more than three orders of magnitude in interbed A-B, more than six orders of magnitude in interbed B-C, and more than seven orders of magnitude in interbed C-D (Barracough and others, 1976; McCarthy and McElroy, 1995). Barracough and others (1976, p. 50) report a range of vertical hydraulic conductivity for these sedimentary units in ten wells, USGS 87, 88, 89, 90, 91, 92, 93, 94, 95, and 96 (figs. 2-2 and 2-3), of from  $1.6 \times 10^{-7}$  to 3.0 m/d. The smallest values of vertical hydraulic conductivity reported for interbed C-D in each of these wells ranges from  $8.0 \times 10^{-7}$  m/d in well USGS 88 to  $2.6 \times 10^{-1}$  m/d in well USGS 91. The largest values of vertical hydraulic conductivity reported for interbed C-D in each of these wells ranges from  $6.1 \times 10^{-5}$  m/d in well USGS 89 to  $9.3 \times 10^{-1}$  m/d in well USGS 95. The greatest range of vertical hydraulic conductivity reported for interbed C-D in any well,  $8.0 \times 10^{-7}$  to  $1.3 \times 10^{-1}$  m/d, occurs in well USGS 88 (Barracough and others, 1976). These ranges of hydraulic conductivity probably reflect centimeter-scale changes in lithology, from clay- to gravel-sized clasts, within the surficial sediment and sedimentary interbeds A-B, B-C, and C-D.

#### 2.4.3.7 Current qualitative understanding of flow in the unsaturated zone at the Idaho National Engineering and Environmental Laboratory

##### 2.4.3.7.1 Vertical flow of local origin

Local infiltration from rainfall, snowmelt, and runoff initially moves downward. If the infiltration is uniform over a large enough area, it can be treated as one-dimensional vertical flow. At many locations, including the SDA, most of the infiltration eventually exits at the soil surface because of evapotranspiration or other mechanisms. Some fraction of infiltration becomes deep percolation that can continue downward until it reaches the aquifer (Cecil and others, 1992; Nimmo and others, 1999). The depth that water must reach for this to happen is described by some as the maximum depth at which plant roots are active. At most locations on the INEEL, this depth is probably a few meters. In the disturbed area of the SDA, where grasses are the dominant vegetation, the depth of active roots is about 2 m and this is within the surficial sediments.

In the surficial layers, infiltrating water can move quickly down to some depth, perhaps a few centimeters or meters, depending on macropores and soil layers, and

then move more slowly from that level onward (Shakofsky and Nimmo, 1996). The travel time for movement from the land surface to the top of the first basalt layer may range from a few days to a few years.

At the interface of the basalt, water accumulates until the sediment is wet enough to allow breakthrough into one or more fractures, or until the wetted portion of the interface broadens enough to include the entrance to a fracture that forms an effective downward conduit. Sometimes this causes perching just above the basalt interface (Bishop, 1996).

Through fractures in the basalt, water can move faster than it typically moves through the sediments, though only if the fractures are adequately filled. When much water is available to fill the fractures, the travel time through an effectively saturated basalt layer tens of meters thick could be from days to weeks (Dunnivant and others, 1998). Flow through unsaturated fractures is likely to be much slower (Tokunaga and Wan, 1997; Su and others, 1999).

When downward flow through basalt fractures reaches a sedimentary interbed, the flow can move in various ways that are poorly understood at present (1999). This flow, like flow through the surficial layers, can be a combination of diffuse and preferential flow. It is probably slower than flow through fractured basalt. Because interbed characteristics are different from those of the surficial sediment and because evapotranspiration at a basalt-interbed interface is insignificant, the flow at these interfaces is in some ways very different from flow through the surficial sediment. Evidence of perching of water in and near interbeds suggests that, in at least some circumstances, the interbeds conduct downward flow markedly more slowly than the basalts. Whether the interbeds on the whole act more as barriers to downward flow or as zones of substantial preferential flow is not yet clearly known.

##### 2.4.3.7.2 Combined vertical and lateral flow

Large amounts of infiltration episodically occur in features near the SDA such as the BLR, spreading areas, and runoff-collecting depressions. Downward flow is likely to be fast and voluminous compared with flow generated only from local precipitation. Thus, travel times to the aquifer below these features are probably much shorter than where infiltration occurs in smaller amounts. The downward flow rate below these features is controlled by the hydraulic conductivity of some layer, most likely in the surficial sediments. Where adjacent layers within the unsaturated zone differ in conductivity, especially at the interfaces between basalt and sediments, lat-

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eral flow and possibly perching occurs; therefore, a two- or three-dimensional model rather than a one-dimensional model is necessary. Some of the perching that occurs near the SDA could result from water that has moved laterally from a wetter area. This additional water could then reduce the vertical travel times to less than it would be for only locally generated flow. Another possibly relevant process is that lateral preferential flow in perched layers could entrain contaminants in the subsurface and move them horizontally to places where vertical travel times are less than those at the SDA.

### 2.4.3.8 Summary of unsaturated-zone flow

Flow behavior in the unsaturated-zone basalts and sedimentary interbeds and the interfaces between them is not well enough understood to quantitatively predict on the basis of widely accepted theory its effect on the spread of contamination. Flow processes routinely occurring at the INEEL in the unsaturated zone cause both vertical and horizontal flow, and create flow impediments as well as preferential pathways. It is known that some types of behavior of great importance to contaminant transport (perching) are known to occur, and others (significant upward flow from interbeds) are unlikely or impossible. Whether many other types of flow behavior, such as unstable fingered flow in interbeds, occur is unknown.

## 2.4.4 Flow in the saturated zone

The SRP aquifer is composed of interbedded basalt and sediment of Quaternary age. Water in the aquifer generally moves from the northeast to the southwest, principally through interflow zones in the basalt. Depth to water at the SDA is about 180 m. Saturated thickness of the aquifer is unknown at this location, but estimates based on differing geohydrologic criteria range from about 75 to 340 m (Robertson, 1974; Morse and McCurry, 1997; Anderson and Liszewski, 1997). Water-level gradients in the vicinity of the SDA area were about 0.6 m/km in 1995 (Bartholomay and others, 1997). Water levels in this area are affected by periodic infiltration of streamflow from the BLR and spreading areas. Barraclough and others (1976) suggested that local reversals in water-level gradients may occur at the SDA during periods of high infiltration.

The number and ages of basalt flows in the aquifer at the SDA are uncertain because few basalt cores have been obtained from this area. Correlations made by Anderson and others (1996) between corehole C1A and well OW-2 (fig. 2-2), located northeast and southwest of the SDA, respectively, suggest that the aquifer is made up of at least

10 basalt-flow groups, each composed of one or more basalt flows that range in age from about 600 thousand to 1.8 million years.

Porosity and hydraulic conductivity of the basalts range from very small in dense portions of basalt flows to extremely large in the fractures, rubble, and scoria between their contacts. Vertical hydraulic conductivity of individual basalt flows, even in the dense basalts, is greatly enhanced by the presence of vertical fractures that penetrate the flows. Vesicular zones occur near the tops and bottoms of basalt flows. The vesicles commonly are not interconnected and, although the porosity of these units can be large, the hydraulic conductivity can be small.

Ackerman (1995, p. 9 and 10) reported a range of probable values for effective porosity of the aquifer of from 0.10 to 0.25. This range is based on numerous studies, including measurements of more than 1,500 core and outcrop samples (Bishop, 1991; Knutson and others, 1990, 1992). Porosity of individual basalt flows generally is least in dense basalt and greatest in vesicular basalt. Although difficult to measure, the largest porosity occurs in fractures, rubble, and scoria between individual basalt flows. Collectively these porosities are referred to as effective porosity. For analysis of steady-state flow and advective transport in the eastern SRP (Ackerman, 1995), the calibration value for effective porosity was 0.21.

The SDA lies within an inferred vent corridor near the Arco-Big Southern Butte volcanic rift zone. Measurements from 32 closely spaced wells that penetrate similar inferred vent corridors at the Idaho Nuclear Technology and Engineering Center (INTEC) (fig. 1-1), indicate that saturated hydraulic conductivity in such areas can vary three to five orders of magnitude within distances of 150 to 300 m (Anderson and others, 1999). The effective hydraulic conductivity of basalt flows in the SRP aquifer at the INEEL ranges from about 0.003 to 9,800 m/d (Anderson and others, 1999). At the SDA, measured hydraulic conductivity ranges from about 0.003 to 700 m/d. Hydraulic conductivity of less than about 30 m/d generally is attributed to the effects of feeder dikes, thick tube-fed pahoehoe flows, and altered basalt. Hydraulic conductivity of more than about 30 m/d generally is attributed to the effects of thin, tube-fed pahoehoe flows; shelly- and slab-pahoehoe flows; and scoria, spatter, and ash. Hydraulic conductivity from eight wells that trend northwestward along the southern boundary of the SDA ranges from about 0.003 to 0.3 m/d, among the smallest measured hydraulic conductivities at the INEEL. These values probably reflect the presence of concealed dikes associated with at least four inferred volcanic vents

in the unsaturated zone at this location (Anderson and Liszewski, 1997, p. 10). If dikes are present in this area—a speculation also reported by Burgess and others (1994)—they likely extend vertically from beneath the base of the aquifer to the top of interbed C-D. If the small hydraulic conductivity in wells along the southern boundary of the SDA is related to concealed dikes, water and wastes in this area must go around or through the zone of dikes. Although regions of similar hydraulic conductivity in the aquifer at the SDA can be delineated using existing data (Magnuson and Sondrup, 1998), these data may not be sufficient to describe the many significant local variations that are thought to occur within vent corridors (Anderson and others, 1999).

Magnuson and Sondrup (1998) assumed an aquifer thickness of about 75 m for simulations of saturated flow at and near the SDA. They reported that this estimate was originally developed from Robertson (1974) and has been used extensively since that time. Many different methods have been used to estimate the thickness of the aquifer at specific sites at the INEEL since the study by Robertson (1974). These methods include geophysical surveys, studies of basalt cores, and measurement of permeability, porosity, and water temperature (Mann, 1986; Whitehead, 1992; Anderson and Liszewski, 1997; Morse and McCurry, 1997). Estimates based on these methods range from about 120 to more than 360 m. Morse and McCurry (1997) estimated an aquifer thickness of about 137 m in well C1A, about 0.5 km northeast of the SDA. This estimate, which may be the best estimate for the SDA, is based on the depth to the top of altered basalt, and a corresponding decrease in permeability and porosity, and a change from convective to conductive heat flow.

According to Barraclough and others (1976), local ground-water altitude, gradients, and associated rates and directions of flow are affected by four composite recharge sources: (1) regional flow from the northeast, (2) infiltration from local precipitation, (3) infiltration from the BLR, about 3 km northwest of the SDA, and (4) infiltration from the INEEL spreading areas, about 1.5 km southwest of the SDA, which periodically contain overflow from the river. According to Barraclough and others (1976), the predominant influence appears to be the spreading areas. During years of larger discharge and associated diversions to the spreading areas, such as 1965, 1969, 1983, and 1984 (Bartholomay and others, 1997), local gradients at the SDA may be northeastward (Barraclough and others, 1976). During years of small or no discharge and diversions, such as 1988 to 1994, local gradients may be southwestward. Ground-water velocities in the southern part of the INEEL range from about 2

to 7 m/d and average about 3 m/d (Ackerman, 1995, p. 11; L.D. Cecil, USGS, oral commun., 1999). Ground-water velocities at the SDA probably are within this range.

## 2.5 Contaminant transport

### 2.5.1 Source-term description

Transport of radioactive contaminants of potential concern (COPC's) at the SDA is directly related to the type of contaminant source, the chemical form of the contaminants, and the chemical environment where the contaminants reside. The exact chemical form of transuranic waste originally disposed of at the SDA is unknown and it is possible that the chemical environment in the subsurface has changed since emplacement of the waste. The contaminant sources at the SDA and surrounding area include both point sources and nonpoint sources.

#### 2.5.1.1 Point sources

Radioactive waste has been buried at the SDA since 1952. From 1952 to 1970, low-level and transuranic radioactive wastes were buried in pits and trenches excavated into the surficial sediment. Wastes sometimes were dumped randomly into the pits and compacted, which could have damaged containers. Since 1970, burial of low-level radioactive waste has continued and transuranic waste has been stored on above-ground asphalt pads in retrievable containers. Between 1952 and 1986, more than 170 thousand m<sup>3</sup> of low-level and transuranic waste containing about 9.5 million Ci of radioactivity was buried at the SDA (Davis and Pittman, 1990, p. 2). Of this 9.5 million Ci, about 0.5 million Ci was transuranic radioactive waste (Roland Felt, DOE, oral commun., 1999). An inventory of annual amounts of 38 radioactive contaminants that were buried was provided by Becker and others (1998, table 4-1).

The chemical form of the buried transuranic waste is unclear; however Navratil (written commun., 1996) indicated that the major plutonium residue processes at Rocky Flats, Colorado consisted of dissolution, ion exchange, precipitation, fluorination, and reduction. Plutonium tetrafluoride (PuF<sub>4</sub>), a finely divided powder, was handled in the last two processes. Because the powdered PuF<sub>4</sub> easily contaminates items such as plastics, paper, gloves, and filters, it is reasonable to conclude that waste buried at the SDA contained plutonium in the form PuF<sub>4</sub>. Other forms of plutonium also may be buried at the SDA. The transuranic waste buried at the SDA is in a variety of different physical forms that include containerized sludge, assorted

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solid wastes, empty contaminated drums in cardboard boxes, sewage sludge, nitrate salts, depleted uranium waste, and secondary sources such as contaminated asphalt and soil (Becker and others, 1998, p. 3-38 to 3-42). Oxygen in the unsaturated zone at the SDA may react with organic material buried in the pits and trenches, which may create localized reducing conditions and environments where some forms of plutonium are more mobile than others. Stability and movement of various plutonium species are discussed in chapter 4 of this document.

The waste at the SDA is packaged in a variety of containers, such as steel boxes, concrete casks, steel drums, plywood boxes, and other containers (Becker and others, 1998, p. 6-2). Breaching of these containers or diffusion of contaminants through the container walls allows contaminant release that can be called point-source contamination. Although this definition does not precisely follow the definition of point source contamination, the nonsystematic release of contaminants in a small area and the release by historic waste disposal methods combine to produce many individual point sources of contamination in close proximity. These point sources can be very diversified in actual contaminant composition. Once released from their containers, the contaminants can be transported away from the release sites in a variety of ways, which are discussed in other parts of this document.

### 25.1.2 Nonpoint sources

Concentrations of selected radionuclides that are slightly larger than background concentrations have been detected in surficial sediments near the SDA (Markham, 1978; Beasley and others, 1998, p. i). For example, the range of  $^{239,240}\text{Pu}$  concentrations near the SDA is from about 50 pCi/m<sup>2</sup> to greater than 600 pCi/m<sup>2</sup>; however, concentrations in 11 of 19 samples ranged from 270 to 650 pCi/m<sup>2</sup> (Beasley and others, 1998, fig. 15). These radionuclides in the shallow subsurface are a potential source of contamination to the unsaturated zone and, ultimately, to the eastern Snake River Plain aquifer. The concentrations of these radionuclides at the SDA are sufficiently large to be measured with conventional methods, but at distances of more than about 2 km from the SDA, mass spectrometry would be required to detect the concentrations (L.D. Cecil, U.S. Geological Survey, oral commun., 1999). In the layered unsaturated zone beneath the SDA, these contaminants potentially could be transported by a variety of natural preferential-flow processes that include, but are not limited to, macropore flow, funneled flow, or unstable flow. Chemical, biological, or physical means of mobilizing or retarding contaminants could affect contaminant migration. The specific means

of mobilizing or retarding contaminants and the types of flow that could transport the contaminants are discussed in other parts of this document. One implication of the widespread distribution of slightly larger than background concentrations of radionuclides in surficial sediments is that extreme care should be exercised during drilling operations to avoid dragging contaminants into the deeper subsurface.

## 25.2 Summary of geochemical processes at the Subsurface Disposal Area

Several studies of geochemical processes at the SDA have been done. Olmstead (1962) and Robertson and others (1974) described the chemical and physical properties of ground water beneath the INEEL, which included analyses of ground water near the SDA. Knobel and others (1997) delineated the geochemical reactions in the SRP aquifer at the SDA and other locations at the INEEL. Rightmire and Lewis (1987b) examined the geochemistry of the unsaturated zone at the SDA. C.T. Rightmire and B.D. Lewis (USGS written commun., 1995) examined the effect of the geochemical environment on radionuclide migration in the unsaturated zone at the SDA. Rawson and Hubbell (1989) and Rawson and others (1991) studied the geochemical controls on the composition of soil pore water in the unsaturated zone beneath a mixed-waste disposal site at the SDA to determine the extent of radionuclide migration from the disposal site. Cleveland and Mullin (1993) studied the speciation of Pu at various oxidation states and Am in ground water at the SDA and found that the solubility of Am was generally low in all cases. Their results also indicated that low-oxidation-state Pu was generally insoluble in water from the SRP aquifer, was more soluble in the perched water, and could be leached from waste and ultimately reach the aquifer. Dicke (1997) examined the distribution coefficients and contaminant solubilities for the WAG-7 Baseline Risk Assessment. A generalized overview of information from these and other reports is given in the following section.

### 25.2.1 Lithology and mineralogy

Understanding the lithology and mineralogy of sediment and rock below the SDA is important in characterizing the materials present that may inhibit radionuclide migration. Several studies have examined the lithology and mineralogy of surficial sediment, sedimentary interbeds, and vesicle- and fracture-infill material at the SDA. Core sample descriptions, thin-section observations, and observations made using scanning electron microscopy for selected cores were presented by Rightmire (1984)

and Rightmire and Lewis (1987a). Voegeli and Deutsch (1953) and Nace and others (1956) presented the cation exchange capacity (CEC) and mineralogy of surficial sediment. Additional data on surficial sediment at and near the SDA were presented as follows: Barraclough and others (1976)—CEC, mineralogy, and grain-size distribution data; Rightmire and Lewis (1987b)—CEC, mineralogy, and grain-size distribution data; Bartholomay and others (1989)—mineralogy and grain-size distribution data; Liszewski and others (1997)—mineralogy and grain-size distribution data; and Liszewski and others (1999)—chemical composition data. Grain-size distribution and CEC data from samples from sedimentary interbeds and vesicle- and fracture-infill material were presented by Barraclough and others (1976) and Rightmire (1984). Mineralogy of sedimentary interbeds and/or vesicle- and fracture-infill material was presented by Barraclough and others (1976), Rightmire (1984), Rightmire and Lewis (1987b), Bartholomay and others (1989), Burgess and others (1994), and Reed and Bartholomay (1994).

Typical basalt geochemistry, sediment grain lithology, and mineralogy were discussed earlier in this chapter. Analyses of distribution coefficients for several radionuclides for basalts indicate little exchange between basalts and water; thus, basalt probably is not much of a factor in preventing radionuclide migration (Dicke, 1997; Collelo and others, 1998). Pace and others (1999) found that vesicle and fracture infill material produced the largest strontium distribution coefficients of any material at the INEEL and were the most likely sites for sorption or exchange with radionuclides in solution. A more detailed discussion on the use of distribution coefficients for predicting contaminant transport is given in chapter 5.

## 2.5.2.2 Water chemistry

Several water properties can affect the solubility of radionuclides; these properties will be discussed in more detail in chapter 4. To evaluate water properties, water chemistry needs to be known. Water-chemistry data from the BLR at Arco, Idaho, upstream from the SDA, were given by Rightmire and Lewis (1987b) and Bartholomay (1990b). The surface water is a calcium bicarbonate type that includes small amounts of magnesium and sulfate. Water-chemistry data for perched ground water above the C-D interbed were presented by Barraclough and others (1976), Rightmire and Lewis (1987b), Hubbell (1990), Knobel and others (1992a), Tucker and Orr (1998), and Bartholomay (1998). Water chemistry data for several suction lysimeters at the SDA were presented by Laney and others (1988) and L.C. Hull, BBWI (unpub. data, 2000). The results of a search for data on quality of

perched water at the SDA along with a list of references for the data was presented by Becker and others (1998, table 4-7). Rawson and others (1991) determined major ion concentrations of soil pore water samples. Piper plots indicate that water in the shallow boreholes was either a sodium chloride or sodium bicarbonate type and water in the deeper boreholes was a sodium chloride type (Rawson and Hubbell, 1989, p. 245). C.T. Rightmire and B.D. Lewis (USGS written commun., 1995) described possible chemical reactions that could take place if infiltrating water reacts with the available minerals.

Saturation indices (SI) calculated from the analyses of perched-water at the SDA indicate that many of the mineral phases with which these water samples were oversaturated were those with large exchange capacities: zeolite minerals (laumontite, phillipsite, wairakite) and clay minerals (calcium smectite and illite) (Rightmire and Lewis, 1987b). As discussed previously, illite was the main exchange mineral present. Saturation indices of soil pore water samples show oversaturation with respect to quartz, chalcedony, cristobalite, calcite, talc, tremolite, iron oxides, hydroxides, oxyhydroxides, and a wide variety of aluminous phyllosilicates and tectosilicates (Rawson and Hubbell, 1989, p. 245-246).

Water-chemistry data for water from eight wells completed in the SRP aquifer at and near the SDA were presented by Mann and Knobel (1988), Knobel and others (1992a), and Liszewski and Mann (1993). Water chemistry data for water from six other wells at the SDA were presented by Burgess and others (1994). Concentrations of selected radionuclides in ground water at the SDA were reported by Barraclough and others (1976), Knobel and Mann (1988), and Knobel and others (1992a). The results of a search for data on ground water at the SDA were presented by Becker and others (1998, tables 4-11 through 4-13). The water samples from the aquifer at the SDA generally are a calcium magnesium bicarbonate type (Knobel and others, 1997), which is typical of most water in the SRP aquifer. Water samples from three wells at the SDA were slightly enriched with sodium plus potassium, and water from two wells were slightly enriched with chloride (Knobel and others, 1997, p. 20).

Median SI values for the carbonate minerals calcite, and dolomite indicate that the SRP aquifer water samples from the SDA are supersaturated with respect to these minerals; therefore, precipitation of the minerals is the only thermodynamic possibility. The median SI values with respect to fluorite; the sulfate minerals celestite and gypsum; and the silicate minerals diopside, clinoenstatite, and olivine indicate undersaturation and, hence, dissolution of these minerals, if present in the aquifer. Median SI

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values with respect to crystalline quartz and cryptocrystalline chalcedony indicate that they are stable in the solid phase; silica glass is unstable, but near equilibrium (Knobel and others, 1997, p. 20–24).

In a mineral stability diagram for the potassium system, all water chemistry data for samples from wells at the SDA plotted in the kaolinite stability field, which indicates that microcline, muscovite, and gibbsite are unstable solid phases and would react with water to form kaolinite. In similar diagrams for the plagioclase system, data for water associated with anorthite plotted in the calcium montmorillonite field, and data for water associated with albite plotted in the sodium montmorillonite field (Knobel and others, 1997, p. 24–33).

### 2.5.3 Description of contaminant transport processes

#### 2.5.3.1 Solute transport

Solute transport is described further in chapters 4 and 5 of this report. In addition to hydrologic processes such as advection, dispersion and diffusion, chemical processes will strongly affect the transport of most radionuclides by controlling or limiting their aqueous concentrations. Redox-sensitive aqueous-phase constituents and adjoining redox-sensitive minerals (such as Fe(II)-rich biotite or organic carbon) may affect the solubility of radionuclides such as U, Pu and Np by controlling their oxidation state. Generally, the nuclides under higher oxidation states, such as U(VI), Pu(VI), Pu(V), and Np(V) will be more soluble than the equivalent nuclides under lower oxidation states (for example U(IV), Pu(IV), and Np(IV)). The major ion composition of the waters may also affect the solubility/mobility of the nuclides by providing radionuclide complexing agents (such as carbonate, sulfate, and phosphate ions). The effect of pH on the hydrolysis of the radionuclides will also be an important control on their solubility/mobility. Similarly, the mineralogy and composition of the solids contacting the contaminated waters will also be important, not only through their controlling effects on the redox state, complexing ion concentrations and pH of the waters, but also by providing surfaces on which the radionuclides may sorb. Minerals such as Fe-oxyhydroxides (of which goethite will be the most predominant), calcite, clays, solid-organic matter can be expected to be strong sorbents for various radionuclides depending on the aqueous speciation of the nuclides and on the speciation of the sorbing surfaces, which will often be pH dependent. The presence or formation of radionuclide minerals may also control their aqueous concentrations, particularly in the source areas.

A deterministic model of radionuclide transport requires a large amount of information on (1) the chemistry, mineralogy and amounts of solids and solid surfaces, (2) the chemistry of the various contacting waters, and (3) the thermodynamics and kinetics of the various chemical processes of importance. Because of the difficulty of characterizing, to a sufficient level of detail, all the relevant information, solute transport models often adopt a more empirical approach; they commonly assume that the retardation of a contaminant of interest relative to the movement of the water can simply be described as constant and uniform throughout the simulation domain. In other words they use the  $K_d$  approach.

#### 2.5.3.2 Colloid transport

Transport of radionuclides beneath WAG-7 could be enhanced by association with colloids and larger particles capable of being transported by water. Colloids can be rock and mineral fragments, mineral precipitates and weathering products, macromolecular components of dissolved organic carbon such as humic substances, biocolloids such as micro-organisms, and microemulsions of nonaqueous phase liquids.

Actinides can form colloids by hydrolysis and precipitation. These reactions are affected by pH, the concentration of complexing agents, actinide concentration, and oxidation state. For example, Am solubility is at a minimum at pH 8. Both Np(IV) and Pu(IV) are much less soluble than other oxidation states of either element. Colloid as used in this section includes any non-aqueous phase state.

The mobility of colloids in the subsurface is controlled by the stability of the colloids in ground water, chemical interactions between colloids and immobile matrix surfaces, and hydrological and physical factors. Particles can be removed through mechanical filtration by smaller pore spaces. Changes in aqueous chemistry can cause aggregation of colloidal particles, or if electrostatic and London-van der Waals forces are present, attachment of particles to immobile surfaces. Colloids can be destabilized by increases in ionic strength, which result in compression of the double layer surrounding particles, or by pH-induced changes in surface charge.

There is evidence for transport of actinides as colloids during batch and column experiments (for example, Ramsay, 1988; Champ and others, 1982). Actinides are associated with a wide range of particle sizes, from 500 molecular weight to clay sized and larger. In the batch and column experiments, actinides often eluted from columns at much faster rates than would have been predicted from equilibrium sorption of aqueous species. Generally, the



form of the actinide eluting from the columns was not determined, so whether the fast elution was facilitated by transport as a colloid or transport as an aqueous complex is undetermined. However, in some experiments there was enough secondary evidence to implicate transport as a colloid.

Colloid facilitated transport of actinides in the field also has been documented. Pu and Am were found beneath and downgradient from waste-disposal beds at Los Alamos, NM at distances that indicate colloid facilitated transport (Nuttall and others, 1991). Arguments, although controversial, suggest that there also is evidence for migration of Pu as a colloid at the Nevada Test Site (Kersting and others, 1999). Isotopic ratios show that the Pu, measured in ground water came from a particular underground explosion 1.3 km away from the blast site. The 30-year travel time is consistent with the travel time for ground water in the area. U associated with colloidal kaolinite and silica has been detected in ground water from Australia (Payne and others, 1992).

Colloids are present in the basalt fractures at the INEEL and also have been measured in ground water. The tighter packed interbed sediments beneath the SDA could inhibit transport of colloids by filtration; however, colloid transport through the coarser fractures is a possibility. Detections of Pu and Am in the interbeds at the INEEL may indicate colloid-facilitated transport.

## 2.6 Summary

Diverse hydraulic processes control contaminant transport through the 200-m-thick unsaturated zone in the southwestern portion of the INEEL. The interbedded basalts and sediments that comprise this unsaturated zone are structurally complex in terms of both preferential-flow paths and layers that contrast in properties such as thickness, permeability, and porosity. The permeability of the fractured basalts is generally much larger than that of dense basalts and sediments.

The unsaturated zone at the INEEL site is complex in ways that have not been much investigated in other settings. The unsaturated zone near the SDA is about 200 m thick, and comprises granular media as well as fractured rock. Its uncommon features, relative to the most widely studied earth sites, include great thickness, severe contrasts between layers in the form of fractured igneous rock alternating with unconsolidated sediments, intense stratification of the media, and large nonuniform inputs of water over the land surface.

Vertical water movement through the basalt-sedimentary sequence is largely controlled by the hydraulic

conductivity of fractures in the basalt flows and by the lower hydraulic conductivity of some of the interbeds. The interfaces between sediments and basalt may have significant additional effects, both from the commonly expected phenomena of flow through contrasting layers in the unsaturated zone, and also from mechanisms peculiar to the combination of sediments and fractured rock.

The basalts and sedimentary layers and the interfaces between them are not well enough understood with respect to their unsaturated-zone flow behavior to predict on a theoretical basis the effect of unsaturated-zone flow on the spread of contamination. Various processes routinely operate in the unsaturated zone in this area to cause vertical and horizontal flow. Some relevant types of behavior (for example perching) are known to occur, and others (for example significant upward flow from interbeds) are unlikely or impossible, but there are many that may or may not occur (for example unstable fingered flow in interbeds) with an unknown net effect on contaminant transport.

Seasonal streamflow, local runoff, and snowmelts routinely generate large quantities of infiltrating water for periods of days or weeks. Particular concerns arise regarding seasonal diversion of water from the BLR to spreading areas located as close as 1 km to the SDA. Short-term, large-volume infiltration may cause rapid, long-range transport phenomena.

Vertical flow occurs mainly through highly conductive fractures. As a result of substantial surface inputs of water, some of the water perches in or on the interbeds, or on basalt flows of lesser effective vertical conductivity. The perched zones may persist for a few months or longer, until horizontal and vertical flow spreads them out enough to leave the porous materials unsaturated. The behavior and composition of perched water observed since the 1970's suggest that this water may sometimes move within the unsaturated zone at unusually high flow rates and volumes. These effects can severely affect contamination in various ways, including fast horizontal transport.

Certain features of basalt flows, volcanic rift zones, and sedimentary interbeds may constitute hydraulic pathways for the preferential movement of water in the unsaturated zone and in the aquifer (Anderson and others, 1999). Preferential-flow paths associated with subsurface features can transport water and contaminants horizontally to adjacent regions or vertically to the aquifer far sooner than might be predicted based on bulk medium properties and Richards equation. An important secondary feature of preferential flow is that the interaction with contaminants is effectively limited to a fraction of the subsurface medium, limiting adsorption and other attenuating



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processes. The equivalent-granular-medium approach is essentially the only fully practical means of treating preferential flow over large scales that has been widely applied, though it has significant limitations.

Rubble and fracture zones provide the main conduits for flow in the saturated zone. The effective hydraulic conductivity ranges from about  $3 \times 10^{-8}$  to  $8.4 \times 10^{-2}$  m/s. The aquifer is assumed to be anisotropic, with estimated ratios of horizontal to vertical basalt permeabilities ranging from about 3:1 to 300:1 (Barraclough and others, 1976; Magnuson, 1995; Magnuson and Sondrup, 1998).

Key issues related to subsurface contaminant transport include (1) travel times to and within the aquifer, both average or typical values and the range of values to be expected, and (2) modes of contaminant transport, especially adsorption processes. Some features promote transport of radionuclides, for example preferential flow, irregularity of layer interfaces, flooding, and perched water. Some features inhibit transport of radionuclides, for example layer contrasts, fineness of grains in interbeds, thickness of the unsaturated zone, relatively low average precipitation, and low porosity of many of the media, such as basalt between fractures.

## 3.0 Task 1: Review of radionuclide sampling program and available data at the Subsurface Disposal Area

### 3.1 Introduction

This chapter is divided into five parts. The first part is a review of the methods used by the site contractor to monitor for and report detections of selected radionuclides in the environment near the Subsurface Disposal Area (SDA) (section 3.2). The second part is a review of the site contractor's sampling methodology (section 3.3), analytical methods (section 3.4), and quality assurance and quality control protocols (section 3.5). The third part is a review of data from Lockheed Martin Idaho Technologies, Inc. (LMITCO) for reported detections of radionuclides in the unsaturated and saturated zones at and near the SDA (section 3.6). The fourth part is a brief discussion on the significance of the reported detections (section 3.6.3) and the fifth part is a list of specific recommendations to improve sample collection and analyses, data generation and reporting, and overall quality assurance and quality control for generating data in support of the Waste Area Group 7 (WAG-7) Comprehensive Remedial Investigation and Feasibility Study (RI/FS) (section 3.7). Multiple criteria were considered in evaluating the data. These criteria were (1) statistical significance, (2) isotopic association, (3) consistency of detections with the historical data record, (4) possibility of cross contamination, and (5) adequacy of the sampling program.

Tables referenced in this chapter are located at the end of section 3.7.

#### 3.1.1 Background

Radionuclide data collected in support of the WAG-7 Comprehensive RI/FS will be used to determine the possible release of radioactive contaminants from the SDA and to aid in understanding the fate and transport of radionuclides (Barrie and Haney, 1997). The specific data quality objectives (DQOs) for radionuclide sampling of six perimeter ground-water monitoring wells near the SDA are to (1) detect contaminated ground water moving away from the SDA, (2) determine the lateral and vertical extent of contamination, (3) obtain samples representative of water quality, and (4) determine the ground-water flow direction and velocity. The specific DQOs for four ground-water monitoring wells upgradient in the far field are provided by the DOE (1998, table 4-3). They are to (1) identify contaminant concentrations in the aquifer to

determine whether the contaminants are derived from the SDA or from an upgradient source, (2) delineate the areal extent of the carbon tetrachloride ( $\text{CCl}_4$ ) plume in the unsaturated zone upgradient of the SDA, (3) identify well (M14S, fig. 2.3) to determine background concentrations for long-term monitoring, (4) calibrate fate and transport models that predict plume dimensions and concentrations, and (5) track and discriminate the Test Reactor Area (TRA) and the Idaho Chemical Processing Plant (ICPP) (currently called the Idaho Nuclear Technology and Engineering Center (INTEC)) plumes and evaluate risk from SDA contaminants.

#### 3.1.2 Purpose and scope

The purpose of this review is to determine whether the Department of Energy (DOE) site contractor at the INEEL is using scientifically defensible methods to quantify and monitor selected radionuclides in the environment near the SDA. This review includes the complete measurement process: reporting of data, sample collection and analysis, and quality control of selected radionuclide data. Therefore, each of these areas will be reviewed as they pertain to the criteria outlined in section 3.1. The scope is confined to WAG-7 and includes an evaluation of the methods used by the site contractor to monitor for and report detections of  $^{241}\text{Am}$ ,  $^{232,234}\text{U}$  undivided,  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  undivided, and  $^{230}\text{Th}$ .

### 3.2 Detection limits, statistical screening criteria, and reporting of data

The detection limit currently used by the site contractor for  $^{241}\text{Am}$ ,  $^{232,234}\text{U}$  undivided,  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  undivided, or  $^{230}\text{Th}$  in water is 0.05 picocuries per liter (pCi/L) (Dave Burgess, LMITCO, written commun., 1998). For these selected radionuclides in soils and other solid matrices, the detection limit is 0.003 pCi/g. As discussed in section 3.4, "Analytical Methods," these detection limits are adequate unless questions are raised about reported detections that are at these limits. Improvements in precision for detections at and near the quoted limits can be gained by using isotope dilution mass spectrometry, thermal ionization mass spectrometry, or accelerator mass spectrometry.

Data-reporting criteria outlined by Becker and others (1996) call for reporting detections to the DOE and State of Idaho if an analytical result is equal to or larger than two sample standard deviations ( $2s$ )<sup>1</sup>. Becker and others (1996) further noted that when the above criterion is met, there is a 95-percent confidence that the target analyte has

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been detected. However, the correct application of the statistics of detection (hypothesis testing) according to Currie (1984) and the American Society for Testing and Materials (ASTM) (1996) leads to the conclusion that at 2s, there is a 95-percent confidence that the true concentration is only within a range of plus or minus the associated uncertainty. For example, for the result  $5 \pm 2$ , there is a 95 percent confidence that the true concentration is somewhere between 9 and 1. This does not address the question of detection or nondetection. During an interview of LMITCO personnel, the question of the origin of the 2s criterion was addressed and the consensus was that the 2s criterion was based solely on historical precedence (Kirk Dooley, LMITCO, oral commun., 1998).

Discussion with personnel from LMITCO, DOE, and the State of Idaho indicated that, among these groups, 2s is the agreed-upon reporting criterion for detections; however, hypothesis testing is the most defensible method for determining statistical detection of a target analyte. The following illustration of the application of hypothesis testing to the question of detection for any radionuclide is based on a method outlined by Currie (1984) and Cecil (1989), and is the method recommended by ASTM (1996) for reporting low-level radionuclide data.

In the analysis for radionuclides, laboratory measurements are made on a target sample and a prepared blank. In this discussion, it is assumed that the blank is a so-called well-known blank (Currie, 1984). Instrument signals for the sample and the blank vary randomly. Therefore, it is essential to distinguish between two key aspects of the problem of detection: (1) the instrument signal for the sample must be larger than the signal for the blank before the decision can be made that radionuclides were detected, and (2) an estimation must be made of the minimum radionuclide concentration that will yield a sufficiently large signal before the correct decision can be made for detection or nondetection of the radionuclide. The first aspect of the problem is a qualitative decision based on an observed signal and a definite criterion for detection. The second aspect is an estimation of the detection capabilities of a given measurement process.

In the laboratory, instrument signals must exceed a critical level before a qualitative decision can be made as to whether the radionuclide was detected. As a guide, radionuclide concentrations that are equal to 1.6 times their associated sample standard deviation meet this criterion. At 1.6s, there is about a 95-percent probability that the correct decision, not detected, will be made. The mul-

tiplier on the sample standard deviation, 1.6, varies with background counts and is based on a measurement process that includes data for a well-known blank. Given a large number of samples, as many as 5 percent of the samples with measured concentrations larger than or equal to 1.6s, which were concluded as being detected, might not contain radionuclides. These results are referred to as false positives and are errors of the first kind in hypothesis testing. Hypothesis testing may be applied to individual analytical results to determine detection versus nondetection.

Once the critical level of 1.6s has been defined in reporting data, the minimum detectable concentration may be estimated. Radionuclide concentrations that equal 3s represent a measurement at the minimum detectable concentration. Again, the multiplier on the sample standard deviation, 3, varies with background counts and is based on a measurement process that includes data for a well-known blank. For true radionuclide concentrations of 3s or larger, there is about a 95-percent probability that the radionuclide was detected in the sample. In a large number of samples, the conclusion "not detected" will be made in about 5 percent of the samples that contain true concentrations at the minimum detectable concentration of 3s. These results are referred to as false negatives and are errors of the second kind in hypothesis testing.

True radionuclide concentrations between 1.6s and 3s have larger errors of the second kind. That is, the probability of false negative results for samples with true concentrations between 1.6s and 3s is larger than 5 percent. However, between 1.6s and 3s there may be true radionuclide concentrations in the sample. By equating 1.6s and 3s without discussing the possibilities of a true concentration between 1.6s and 3s, the probability of false negatives is about 50 percent. In other words, if only the 3s minimum detectable concentration is used as a guide, at least 50 percent of the time, true concentrations between 1.6s and 3s will be missed.

LMITCO uses 2s as the criterion for detection versus nondetection and reports the results at 2s and larger as being 95-percent probable for detection. This is not an application of hypothesis testing and not a correct application of detection statistics. By not explaining the associated uncertainties of detection in terms of hypothesis testing, the end user of the data is being misled into thinking that at 2s there is a 95-percent probability that radionuclides have been detected in the sample. Using a one-tailed statistical test that is not based on hypothesis testing only gives a 95-percent probability that the result is within a range that is plus or minus 2s about the analyt-

<sup>1</sup> In this report, "s" represents the sample standard deviation and "σ" the population variance. These two terms are often used interchangeably and thus incorrectly in the references cited.

ical result. This does not answer the question of detection versus nondetection.

The hypothesis testing method of reporting low-level radionuclide data is recommended by the National Institute of Science and Technology, ASTM, the International Union of Applied and Pure Chemistry (IUPAC), and is used by the USGS. Personnel at DOE and LMITCO who are responsible for reporting radionuclide data generally do not use hypothesis testing to determine detection of radionuclides. It is recommended that all personnel at DOE, LMITCO, and the State of Idaho address this issue in reporting detections of radionuclides. If using 2s as the screening criterion for detection is to be continued, then it should be documented in the Quality Assurance Plan (QAP) or Sampling and Analysis Plan (SAP) as an arbitrary DQO that is not based on hypothesis testing.

In reviewing the reporting of detections for radionuclide data, two related problems concerning data validation were discovered. The first involved reporting detections that were apparently orders of magnitude larger than the analytical method detection limits with associated uncertainties as large as 30 percent. The following documentation is offered for illustration. On February 3, 1998, the USGS brought to the attention of DOE inconsistencies in reported detections of  $^{241}\text{Am}$  and  $^{54}\text{Mn}$ . The  $^{241}\text{Am}$  concentration was reported as  $1.03 \pm 0.27$  pCi/L. This concentration is almost two orders of magnitude larger than the method detection limit and the associated uncertainty is almost 30 percent of the concentration. LMITCO personnel were notified, and they subsequently learned that the laboratory had omitted a calibration correction. When the error was corrected, the associated uncertainty was reduced to 0.03 pCi/L. According to personnel from the LMITCO's Sample Management Office (SMO) (Cliff Watkins and Don Koeppen, LMITCO, oral commun., 1998) the calibration correction was missed initially because a Level C instead of a Level B validation had been performed.

The second problem concerning data validation involved reporting detections of isotopes that are highly unlikely to be in environmental samples collected in the aquifer near the SDA. An example is  $^{54}\text{Mn}$ , which was reported as a detection in a water sample collected in October 1997 from a well completed in the regional aquifer. Because this isotope has a 320-day half-life, it should not have been reported as a detection until a complete data validation had been performed. A screen for exotic isotopes such as  $^{54}\text{Mn}$  in the computer program used to construct the data base for WAG-7 could prevent this type of problem. LMITCO personnel were advised of the reported detection of  $^{54}\text{Mn}$ , and they subsequently per-

formed a complete Level A validation and found that  $^{54}\text{Mn}$  should not have been reported as a detection. In the documentation for reporting data generated in monitoring at the SDA, it is stated that for radionuclides, a Level B validation will be performed. In our interview of LMITCO employees, it was discovered that because of budget considerations, a Level C instead of a level B validation had been performed for both of the reported detections cited above. However, a Level A validation was performed when the analyses were called into question (Kathy Falconer, written commun., 1998). The level of data validation should be consistent for all radionuclides.

### 3.3 Sampling methodology

The sampling methodology used for routine monitoring in support of the WAG-7 Comprehensive RI/FS was described by Barrie and Haney (1997). The specific sections by Barrie and Haney (1997) dealing with sampling methodology are section 3, "Sampling Location, Frequency, and Media," section 4, "Sample Identification," section 5, "Sampling Equipment and Procedures," section 6, "Sample Handling, Packaging, and Shipping," section 7, "Documentation," and section 8, "Handling and Disposition of Investigation Derived Waste".

#### 3.3.1 Sampling location, frequency, and media

Barrie and Haney (1997, fig. 1-1) designated six wells as water-quality monitoring sites. These wells are completed in the Snake River Plain (SRP) aquifer and are designated as M1SA, M3S, M4D, M6S, M7S, and M10S (fig. 2-2). The M in the site identifier indicates monitoring well, the S indicates shallow, approximately 190 m below land surface datum (LSD), and the D indicates deep, approximately 250 m below LSD. All of these sites are in the near field—situated close to the SDA. M3S and M7S are upgradient of the SDA; M6S is slightly southeast of the SDA; and the rest are downgradient. These six perimeter wells were installed in 1992 and were first sampled in October 1992. Four additional wells completed in the SRP aquifer were installed in 1998 and are upgradient of the SDA in the far field—situated at a distance from the SDA (Barrie and Haney, 1997, fig. 1.1). These wells, designated as M11S, M12S, M13S, and M14S (fig. 2-3) were first sampled in July 1998. An additional well (USGS 127, fig. 2-3) was completed in 1999 and will be incorporated into the monitoring program. This well also is upgradient from the SDA in the far field.

Perched-water bodies also are monitored at the SDA and water samples from several perched-water wells and

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lysimeters have been analyzed for water quality. In April 1997, a long-term, perched-water-quality program that consists of quarterly sampling was established at the SDA. It is not clear which wells and lysimeters are included in this sampling program; however, radionuclide concentrations in water from several sites have been reported since, on, or before April 1997. These sites include 77-2, 78-1, 88-02D, 89-01D, D-06, D-15, PA-01, PA-02, PA-03, TW-1, USGS 92, W-04, W-06, W-08, W-23, and W-25.

Generally, the locations of the wells in the ground-water monitoring program near the SDA (if well USGS 127 is incorporated) are adequate to meet the DQOs identified by Barrie and Haney (1997, section 2, p. 2-1) and by the DOE (1998, table 4-3, p. 4-22). Sampling the perched-water bodies and the unsaturated-zone water is more difficult because less water is available (sometimes no water is available) for determining the presence of radioactive contaminants. Because of this shortage of sample water, some components of the monitoring program need to be more rigorously defined in the SAP according to the prioritization scheme given by the DOE (1998, table 4-2, p. 4-16). The sample medium for the ground-water monitoring program is "ground water" and the frequency of sample collection is specified in the SAP Table for Chemical and Radiological Analysis (Barrie and Haney, 1997, appendix B, p. B1-B31). The frequency specified in the SAP for the original 6 perimeter ground-water monitoring wells is quarterly for 19 sampling rounds, and annually or semiannually for 6 lysimeters (PA-01, PA-02, PA-03, PA-04, D-06, and TW-1) (fig. 2-2) for 3 sampling rounds. The time period specified in this table is from the third quarter of fiscal year 1993 through the third quarter of fiscal year 1999. The table is very well planned and provides detailed information on what to sample for and the types of analyses to request; however, the table needs to be updated to include information on sampling rounds that follow the third quarter of fiscal year 1999. The table also needs to be expanded to include schedules for the four wells installed in 1998 that were added to the monitoring program (as indicated in DOE, 1998, p. 4-22), the ground-water monitoring well constructed in 1999 (USGS 127), and the perched-water wells and lysimeters (through fiscal year 2004 as indicated in DOE, 1998, p. 4-16).

#### 3.3.2 Sample identification

Barrie and Haney (1997) specified that all samples collected be uniquely identified by a 10-character sample identification code. Barrie and Haney (1997, appendix B)

list the first six characters of this code under the heading Sampling Activity. When field guidance forms and sample labels are generated for the individual sample rounds, the final four characters are established and printed on the forms and labels along with the other six characters. The entire 10-character code is assigned by LMITCO's SMO and a data management system is used to ensure the uniqueness of the sample identification codes. This procedure appears to be adequate for the unique identification of samples. When appendix B is updated to incorporate the additional sampling rounds, wells, and lysimeters, the sampling activity field also should be updated.

#### 3.3.3 Sampling equipment and procedures

Barrie and Haney (1997, section 5 and appendix A) gave detailed instructions for radionuclide ground water sample collection. Section 5 describes site preparation, field measurements, well purging, and well sampling. Appendix A describes field-sampling methods (FSMs) and standard operating procedures as follows: FSM #1, "Well Purging and Sampling with a Submersible Pump", FSM #2, "Decontamination", FSM #3, "Water level measurements with an electronic recorder", FSM #4, "Water level measurement with a steel measuring tape", and FSM #5, "Hydrolab operation". In general, the sampling procedures and equipment are adequate; however, the introduction to section 5 indicates that these procedures apply only to the six perimeter ground-water monitoring wells. The section should specify that these procedures also apply to the other ground-water, perched-water, and lysimeter sites. Appendix A also contains a copy of correspondence providing guidance on the calculation of the amounts of acid or base that can be added to ground-water samples of assorted sizes so that they are in compliance with the Department of Transportation (DOT) shipping regulations. Table 6 of Section 5 also gives guidance on the amount of acid or base to add to ground-water samples; however, David Sill (DOE, oral commun., 1999) indicated that samples for radionuclide analysis submitted to DOE's Radiological and Environmental Laboratory (RESL) have not been acidified sufficiently. To minimize the loss of radionuclides from solution to the container walls, steps should be taken to ensure adequate sample preservation in the field and the laboratory.

#### 3.3.4 Sample handling, packaging, and shipping

Barrie and Haney (1997, section 6, p. 6-1 to 6-3) provided instructions for sample handling, packaging, and shipping. Procedures for these comply with the applicable

Federal regulations specified by Barrie and Haney (1997, p. 6-1) and are adequate to meet program requirements.

### 3.3.5 Documentation

Barrie and Haney (1997, section 7) provided general guidance on documentation procedures, including the controls and disposition of documents and instructions for data entry and corrections. They also identified and described necessary field documents: sample labels, sample tags, chain-of-custody forms, sample logbooks, shipping logbooks, calibration logbooks, field-team leader logbooks, quality assurance plan, field sampling plan and attachments, and the health and safety plan. These procedures and documents generally are adequate to meet the needs of the data acquisition program; however, a few minor deviations from recommended methods were noted and will be discussed in section 3.5 of this report, on quality assurance (QA) and quality control (QC) protocols.

### 3.3.6 Handling and disposition of investigation-derived waste

Barrie and Haney (1997, section 8) defined the various types of waste that could be generated by the sampling program and the storage and disposition of the wastes. The procedures for storage and disposition of the wastes were carefully considered and are adequate for the needs of the program. One potential type of waste, purge water, is addressed in their table 7. On the basis of previous analyses of samples from the perimeter wells M1SA, M3S, M4D, M6S, M7S, and M10S (fig. 2-2), it was determined that concentrations of various constituents (including radiochemical constituents) were within their release limits (either maximum concentration limits (MCLs) or proposed MCLs) and, therefore, need not be containerized. This is a well-designed table, but it should be updated to include the recently added and planned ground-water monitoring wells, the perched-water monitoring wells, and the lysimeters.

### 3.4 Analytical methods and laboratory techniques

The analytical methods used for routine monitoring samples collected in support of the WAG-7 Comprehensive RI/FS were summarized by Baumer and others (1997). The approved analytical method categories listed by Baumer and others (1997) in section 4.1, are to be used unless changes are documented in the site-specific field sampling plan (FSP) (Barrie and Haney, 1997, appendix C, p. C-1 to C-2). No exceptions are noted in appendix C

of the FSP. The approved method categories listed by Baumer and others (1997) are (1) Environmental Protection Agency (EPA) approved methods, (2) American National Standards Institute (ANSI) standard methods, (3) ASTM standards, (4) industry-accepted methods, and (5) methods described in the laboratory statement of work (SOW) prepared by the SMO. These analytical methods are adequate to meet the requirements of the program if the analyzing laboratory is in conformance with the SOW. However, because some reported detections at the method detection limit have been questioned, it would be useful to list the specific analytical method for each constituent in the SAP Table for Chemical and Radiological Analysis (Barrie and Haney, 1997, appendix B). Whenever possible, technical standards adopted by voluntary consensus standards bodies (such as ASTM) should be used for measuring concentrations of radioactive constituents in water and soils (Turner, 1999). Congress, in the National Technology Transfer and Advancement Act (Public Law 104-113) and its Amendments (Collins, 1999; Turner, 1999), has mandated the use of voluntary consensus standards by all Federal agencies.

### 3.5 Quality assurance and quality control protocols

"Each office or laboratory generating data has the responsibility to implement minimum procedures which assure that precision, accuracy, completeness, and representativeness of its data are known and documented. In addition, an organization should specify the quality levels which data must meet in order to be acceptable" (U.S. EPA, 1980, section 1). To ensure that these responsibilities are met, EPA requires that each data-producing organization must have a written QA project plan covering each monitoring or measurement activity within its purview. U.S. EPA (1980) provides (1) guidelines and specifications that describe the 16 essential elements of a QA project plan, (2) recommendations on the format to be followed, and (3) specifications on how plans will be reviewed and approved. The QA and QC protocols employed for WAG-7 are presented in the quality assurance project plan (QAPjP) (Baumer and others, 1997), and by Barrie and Haney (1997, appendix C). Additional documents relating to the contents and preparation of the QAPjP and the SAP are itemized in U.S. DOE (1998, section 4, p. 4-9). The EPA guidance document (U.S. EPA, 1980), particularly the 16 elements that must be considered in the preparation of a QAPjP, was used for evaluating the QA and QC protocols established for the data acquisition program. Following is a list of the 16 elements

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and our evaluation of how completely these elements are addressed in the QAPjP:

**Title page.**—The signatures required in section 5.1.A are satisfactory.

**Table of Contents.**—The most important recommended items are included in the table of contents, which is adequate for the needs of the monitoring program.

**Program Description.**—The recommendations for this section are adequately addressed in the QAPjP; however, Appendix B of the SAP (Barrie and Haney, 1997) needs to be updated to incorporate the additional sampling rounds, wells, and lysimeters to be in compliance with the last paragraph of Section 1.2 of the QAPjP (Baumer and others, 1997).

**Program Organization and Responsibility.**—The U.S. EPA (1980) implies that responsible individuals be identified. Although the QAPjP (Baumer and others, 1997) does not identify these individuals, it does identify the organizational positions that are responsible for the execution of the QAPjP (Baumer and others, 1997). This should be adequate to meet the requirements associated with program organization and responsibility.

**QA Objectives for Precision, Accuracy, Completeness, Representativeness, and Comparability of Measurement Data.**—The requirements of this section are met in section 2, tables 2-1 through 2-12, and section 9 of the QAPjP (Baumer and others, 1997) and are adequate for the needs of the program.

**Sampling Procedures.**—The specific requirements of this section are addressed in the QAPjP (Baumer and others, 1997, section 3, and tables 3-1 and 3-2) and in the SAP (Barrie and Haney, 1997). The sampling procedures specified in these documents are adequate for the needs of the program; however, the sample collection schedules (Barrie and Haney, 1997, appendix B) and sampling locations (Barrie and Haney, 1997, fig. A-1) should be updated to include the additional sampling rounds, wells, and lysimeters.

**Sample Custody.**—The specific requirements of this section are addressed in the QAPjP (Baumer and others, 1997, section 3.4) and in the SAP (Barrie and Haney, 1997, Section 7.1). The chain-of-custody record and the sample label are very well designed; however, it would be desirable to have an exact location (such as latitude and longitude) on the sample label. The other aspects of sample custody are adequate for the needs of the program.

**Calibration Procedures and Frequency.**—The requirements of this section are addressed in the QAPjP (Baumer and others, 1997, section 4.2) and the SAP (Barrie and Haney, 1997, appendix A). The provisions for cal-

ibration and documentation of calibration are adequate for the needs of the program.

**Analytical Procedures.**—Analytical procedures are addressed in the QAPjP (Baumer and others, 1997, section 4.1). Adequacy of the analytical procedures was previously discussed in this document in section 3.4, "Analytical methods (laboratory techniques)."

**Data Reduction, Validation and Reporting.**—The requirements of this section are addressed in the QAPjP (Baumer and others, 1997, sections 6 and 8). The data-reduction and validation methods, including the levels of data validation, are adequately identified in these sections. If the methods are rigorously followed, they should be adequate for the needs of the program. However, as noted in section 3.2, "Detection limits, statistical screening criteria, and reporting of data," there have been inconsistencies in applying data validation levels in the review process.

**Internal Quality Control Checks.**—The requirements of this section are addressed in the QAPjP (Baumer and others, 1997, section 7 and table 2-12), and the SAP (Barrie and Haney, 1997, section 3 and appendix B). The QC identification and definition of QC sample types in the QAPjP (Baumer and others, 1997) are very good. Consideration should be given to the possibility of adding spiked samples to this list. The specific identification of QA/QC samples in the SAP tables (Barrie and Haney, 1997, appendix B) is also very well thought out and is adequate for program needs; however, the tables should be updated to include the additional sampling rounds, wells, and lysimeters.

**Performance and System Audits.**—The requirements of this section are addressed in the QAPjP (Baumer and others, 1997, section 8). As written, the procedures are adequate to meet the needs of the program; however, some concerns regarding laboratory selection and performance were identified in the Task 5 report submitted to DOE earlier in this investigation (U.S. Geological Survey, 1998).

**Preventive Maintenance.**—The requirements of this section are addressed in the QAPjP (Baumer and others, 1997, section 5) and the SAP (Barrie and Haney, 1997, appendix A). The procedures are adequate to meet program needs.

**Specific Routine Procedures Used to Assess Data Precision, Accuracy, and Completeness.**—The requirements of this section are addressed in the QAPjP (Baumer and others, 1997, section 2, tables 2-1 through 2-12, section 9) and are adequate to meet program needs.

**Corrective Action.**—The requirements of this section are addressed in the QAPjP (Baumer and others, 1997,

section 10) and are adequate for the needs of the program if properly implemented. Some reporting of data as identified in U.S. Geological Survey (1998) (see discussion in this section titled "Data reduction, validation and reporting") indicate that implementation of corrective actions may not be complete. This situation needs to be remedied.

**Quality Assurance Reports to Management.**—The requirements of this section are addressed in the QAPjP (Baumer and others, 1997, section 11) and are adequate for the needs of the program.

The QAPjP (Baumer and others, 1997) meets the EPA requirement of having a written and approved QA plan (U.S. EPA, 1980, section 2.3), and, when combined with the SAP (Barrie and Haney, 1997), provides a framework for producing data of known and documented quality that meet the needs of the program. The few exceptions are noted in the above 16 elements.

### 3.6 Reported detections in sediments and water

Data on surficial sediment and sedimentary interbeds will be discussed first because this media is in close proximity to the buried waste at WAG-7. Any radioactive waste that is mobilized from the pits, trenches, or surface soils may move through the unsaturated zone and contact the sedimentary interbeds before reaching the aquifer. Data for ground water from the regional aquifer and from perched zones at WAG-7 will be discussed last.

#### 3.6.1 Surficial sediments and sedimentary interbed data

Sampling of sediments for radionuclide analysis as part of the environmental monitoring program at the SDA began in the 1970's. The results of these studies (numbered 1-6) are summarized below.

(1) 1971–72 drilling program—During 1971–72, six observation wells, called "interior wells," (USGS 91 to USGS 96) were drilled within the SDA to depths of 72 to 92 m below land surface (Barracough and others, 1976). Well locations were selected (fig. 2-2) to provide representative coverage of the burial ground and to avoid drilling into or through any of the buried waste. Four additional wells, called "exterior wells," (USGS 87 to USGS 90) were drilled at locations surrounding the SDA (fig. 2-2) to depths of 191 to 197 m below land surface. Most of the sampling and analyses of drill cuttings and core material were of the surficial sediments (below 0.5 m) and sedimentary interbeds B-C and C-D at depths of about 34 m and 73 m. A total of 58 sediment samples were analyzed; in 27 of these samples concentrations of one or more of the anthropogenic radionuclides  $^{60}\text{Co}$ ,

$^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{140}\text{Ba/La}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$  were statistically positive (defined by Barracough and others, 1976, as exceeding the 2s counting error). Analyses from the samples containing statistically positive concentrations are shown in table 3-1 (Barracough and others, 1976, table II). The largest concentrations of actinides in subsurface samples from the interior wells were from the B-C interbed at the 31- to 34-m sample interval from wells USGS 93 and USGS 96:

$^{238}\text{Pu} 14 \pm 3 \text{ pCi/kg}$

$^{239,240}\text{Pu} 540 \pm 12 \text{ pCi/kg}$

$^{241}\text{Am} 230 \pm 20 \text{ pCi/kg}$

The report by Barracough and others (1976, p. 74–83) contains an extensive discussion of potential artificial contamination of the samples during drilling and sample handling. The authors concluded that the potential for artificial contamination was greatest for the four exterior wells, which were drilled by using the cable-tool method and water as the drilling fluid. Therefore, although some statistically positive analytical results (determined by using the 2s criteria) were obtained for water from wells USGS 87, USGS 89, and USGS 90 (Barracough and others, 1976, table IV), these results may be artifacts of sampling; radioactive contamination (from global fallout or INEEL operations) in the surficial sediment could have been carried downhole and artificially contaminated the subsurface samples. Concentrations of radionuclides in surface soil in and around the Radioactive Waste Management Complex (RWMC) are given in table V of the report by Barracough and others (1976); an extensive 1973 sampling (done by Aerojet Nuclear Company) of surface soils from within the RWMC showed an average  $^{239,240}\text{Pu}$  concentration of 2,300 pCi/kg. The six interior wells were drilled primarily by using dry-air, wire-line rotary techniques, and special precautions were taken to avoid contamination. Barracough and others (1976, p. 87) concluded that there was evidence of migration of anthropogenic radionuclides, including isotopes of Am and Pu, to the B-C and perhaps the C-D interbeds at the SDA.

(2) 1975 core drilling program—In response to the USGS findings above, Aerojet Nuclear Company and U.S. Energy Research and Development Administration (ERDA)/Idaho Operations initiated a new drilling program in 1975 (Burgus and Maestas, 1976). Three wells, USGS 96A, 96B, and 93A (fig. 2-2), were drilled with air, and continuous cores from the top of the first basalt layer to the bottom of the C-D interbed were recovered. These wells were drilled 3 to 15 m from wells USGS 96 and 93, respectively, the wells noted by Barracough and others (1976) to have the greatest percentage of detections and



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the largest concentrations of most anthropogenic radionuclides measured. The wells are in the oldest part of the burial ground. Contamination-control measures included installation of ground-surface protection (clean gravel) around each well, the use of an asphalt cover, a wooden working platform, and other clean protocols to avoid contact with the ground or dust.

Core samples for analysis were selected from the sediment depths (within the B-C and C-D interbeds) at which radionuclides had been detected during 1971–72 and from a shallow sediment zone overlying the basalt at depths of about 10 to 12 m below land surface. Radionuclide analyses were performed at the same ERDA laboratory—INEEL Health Services Laboratory, Idaho Falls—now the RESL, using the same error propagation techniques that were used previously. The method of calculation of uncertainty in radiochemical analyses was consistent through the 1987 sampling reviewed below (Dames and Moore, 1992, p. 32). Navratil (written commun., 1996) noted that analytical procedures (presumably at RESL) were changed in 1985 to decrease the possibility of interferences in Pu analyses (whether this improved procedure was restricted to reanalyses of initially positive samples is not clear). These new analytical procedures are presumed to be those of Sill (1987) and Sill and Sill (1989) and are described in appendix B of Dames and Moore (1992). Data is shown in table 3-2 (Burgus and Maestas, 1976, table 4). At the 2s uncertainty level, none of the results are statistically positive. Burgus and Maestas (1976) suggested that sample contamination may have been a factor in radionuclide concentrations in samples analyzed by Barraclough and others (1976).

The contamination-control measures developed for this drilling program were used in the 1976–77 (Humphrey and Tingey, 1978, p. 18), 1978 (Humphrey, 1980, p. 5) and subsequent drilling programs (through at least 1987) (Dames and Moore, 1992, p. 35) aimed at assessing contaminants in the interbed sediments. Arguments can be made as to the amount of surficial sediment that would have to be added to a sample to yield a measured concentration in a subsurface sample (Barraclough and others, 1976, p. 79; Dames and Moore, 1992, p. 33). Such arguments may make contamination seem unlikely as the major factor in a measured concentration. However, as the Dames and Moore (1992, p. 35) report aptly notes, the potential for artificial contamination in the field or in the lab is “an irresolvable issue.” However, the potential for its occurrence in the drilling programs conducted at and near the SDA since 1972 has been minimized by the contamination-control measures discussed in this section.

(3) 1976–77 core drilling program—In this EG&G study, the work of Burgus and Maestas (1976) was continued by the drilling of six additional wells in the SDA and one well in the Transuranic Storage Area (TSA) of the RWMC during 1976–77 (Humphrey and Tingey, 1978). The coring techniques and anticontamination measures used by Burgus and Maestas (1976) were used again here. Samples were collected for radiochemical analysis from the A-B, B-C, and C-D interbeds; in addition, samples were collected at irregular intervals from silt and clay material filling fractures in the basalt and from the basalt itself. Samples were analyzed either by the Allied Chemical Corporation laboratory at INEEL (wells 76-1, 76-2, 76-3, fig. 2-2) or by DOE RESL (wells 76-4, 76-4A, 76-5, 76-6, 77-2 (fig. 2-2), and all reanalyses). Initial analyses are shown in tables 3-3 and 3-4 (Humphrey and Tingey, 1978, tables 1, 2). Reanalyses of splits from 13 samples (from the 15 samples showing the presence of one or more radionuclides (at 2s level) upon initial analyses) are shown in table 3-5 (from Humphrey and Tingey, 1978; table 5); the top number is the concentration measured initially, and the numbers below are the concentrations measured during reanalysis. Upon reanalyses, only one statistically positive result was obtained—that for  $^{90}\text{Sr}$  from the top of the CnD interbed at well 76-1. Samples had been ground, sieved, and blended prior to the initial analyses, and splits (10 g) for reanalyses had been removed from the same container. The analytical differences between the first and second analyses may reflect sample heterogeneity problems. This seems unlikely, however, given the 10-g aliquot size, unless contamination was caused by (1) “hot particles”—discrete particles of high specific radioactivity in an otherwise uncontaminated matrix, (2) contamination introduced (despite stringent laboratory protocols) into the first-round samples in the laboratory, or (3) unaccounted-for uncertainty terms, the cumulative effect of which is significant at these extremely low radioactivity levels.

Reanalyzing separate aliquots from a given core sample and looking for repeated statistically positive results qualitatively helps to rule out false positives. When the first result is statistically positive and the second is not, the discrepancy could be caused by an initial false positive, laboratory contamination during the initial analysis, an inhomogeneity problem, or a false negative during the second analysis; the situation here remains ambiguous. On the other hand, if the statistically positive analysis is repeatable, confidence in its accuracy is improved. The strategy is to reanalyze only initially positive samples to rule out false positives; the selective reanalysis approach

may be justified for actinides because of the cost and effort required for such measurements.

(4) 1978 core-drilling program—In 1978, three new wells were drilled in the SDA, and core samples were collected for analysis at RESL (table 3-6) (from Humphrey, 1980; table 1). Split samples from each selected depth were analyzed. In addition, new samples from the cores collected during 1976–77 (Humphrey and Tingey, 1978) were analyzed for  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{90}\text{Sr}$ ,  $^{144}\text{Ce}$ ,  $^{137}\text{Cs}$ , and  $^{60}\text{Co}$  (table 3-7) (Humphrey, 1980, table 2).

For the 1978 samples, there was only one case ( $^{238}\text{Pu}$  in a silt and gravel sample from near the top of the B-C interbed of well 78-5) where there were statistically positive radionuclide concentrations in both of the split samples ( $3.1 \pm 1.0$ ;  $1.7 \pm 0.8$  pCi/kg). None of the radionuclide concentrations in the new 1976–77 samples were statistically positive.

(5) 1979 core-drilling program—Results of this study were presented in an unpublished report (Waste Management Programs Division, Collection and radiochemical analyses of sedimentary interbed samples from the Radioactive Waste Management Complex, Idaho National Engineering Laboratory: 1979, EG&G-2083, February 1984). We were unable to locate a copy of the report, but the findings were summarized and the data tabulated by Dames and Moore (1992). In four samples from the B-C interbed at two wells drilled in the SDA, concentrations of one or more of the radionuclides  $^{241}\text{Am}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{60}\text{Co}$  in both splits were statistically positive at the 2s level.

(6) 1986–87 core-drilling program—In fiscal year 1986, borehole D-02 (fig. 2-2) was drilled to the C-D interbed. Radiochemical analyses by EG&G showed positive results (at the 3s level) for concentrations of  $^{238}\text{Pu}$  in four splits from about 70 m (table 3-8) (from Laney and others, 1988; table 20). Neither these results, nor a single positive result for  $^{239,240}\text{Pu}$  in a sample from the same interval could be confirmed by RESL. It is not apparent if samples were collected from the B-C interbed (Laney and others, 1988).

In fiscal year 1987, borehole TW-1 (fig. 2-2) was drilled to the bottom of the C-D interbed within 4.5 m of borehole D-02. EG&G radiochemical analyses of samples from 31-m interval showed consistent (in multiple splits) statistically positive concentrations of  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$  (table 3-8) (from Laney and others, 1988; table 20). Reanalysis of available splits by RESL confirmed these positive results (table 3-9) (Laney and others, 1988; table 21). The largest actinide concentrations reported by the RESL were  $170 \pm 13$  pCi/kg for  $^{239,240}\text{Pu}$ , and  $908 \pm 7.5$  pCi/kg for  $^{241}\text{Am}$ . EG&G radiochemical analy-

ses of samples from the 69- to 69.5-m interval of the C-D interbed at TW-1 showed no positive results (table 3-8) (from Laney and others, 1988; table 20). Laney and others (1988) concluded that isotopes of Am and Pu had migrated to the B-C interbed; results from the C-D interbed were deemed, as in earlier cases, inconclusive.

(7) Discussion of 1971–87 sampling—The shift in the 1986–87 studies to a 3s decision criterion decreased the chances for false positive results. At 2s, the chance of a false positive is about 2.5 percent (or 1 in 40 samples). At 3s, it is about 0.13 percent (or 1 in 769 samples). Table 3-10 A, B, C, and D (modified from Dames and Moore, 1992, tables 5-5 to 5-8), show all results (from both interior wells and background wells (wells USGS 86–90, and wells 76-6 and 79-1) surrounding the SDA) from 1971 to 1987 that were judged to be positive at the 3s confidence level. It is difficult to explain the fact that in Interval 1, 3 to 23 m below land surface (Dames and Moore, 1992), which includes the A-B interbed and the fracture-filling-material sample, there are no repeatable, statistically positive results. This is puzzling because the subsurface zone closest to the buried waste would be expected to have the largest radionuclide concentrations; indeed, there are larger concentrations of  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$  in the surface soils in areas around the SDA due to global and regional fallout (for example, Nevada Test Site, local emissions, and fugitive dust) than in the subsurface of the SDA (Dames and Moore, 1992, p. 72–73). Perhaps this is a reflection of the fewer number of samples in Interval 1 (27 samples) than in Interval 2 (which includes the B-C interbed; 105 samples) and Interval 3 (which includes the C-D interbed; 128 samples).

Table 3-10 lists samples from intervals between 3 and 80 m below land surface within and surrounding the SDA in which statistically positive (at 3s level) radionuclide concentrations were measured. Many discrepancies between the data in the original reports and in subsequent tabulations were found and are noted herein. Tables 3-11, 3-12, and 3-13 focus on the actinides  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$  in the B-C and C-D interbeds. The occurrence of samples with statistically-significant (3s) detections, with or without verification upon reanalysis, are tabulated (tables 3-11, 3-12, and 3-13) and compared with similar information in tables by Navratil (written commun., 1996). Findings from this study are in general agreement with those of Navratil (written commun., 1996); differences largely reflect wells considered in the sampling pool. More statistically-significant (3s), repeatable detections occur in samples from the B-C interbed than from the C-D interbed. Although there are statistically significant concentrations of actinides in samples from the C-D

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interbed, and the evidence for  $^{238}\text{Pu}$  at this depth in well D-02 is strong (table 3-13), the evidence for actinide transport to the B-C interbed is more compelling than that to C-D interbed.

(8) Other approaches in the Dames and Moore (1992) report—The purpose in each of the studies discussed above was primarily to identify statistically positive detections in samples from the subsurface at the SDA. Dames and Moore (1992) compiled and analyzed this data. In this 1992 study, statistical methods were used to determine whether the concentrations in samples from wells within the SDA boundaries were different from the concentrations in samples from background wells outside the boundaries of the SDA. Because the SDA data generally were not normally distributed, nonparametric statistical testing was used. A detailed analysis of the statistical methodologies they used is beyond the scope of this review. In summary, the entire 1971–87 data set without associated uncertainties was tested primarily by using the Wilcoxon Rank Sum test. Results show that in samples from SDA interior wells:

- $^{144}\text{Ce}$  was present in Interval 1 at concentrations statistically larger than background (note: the maximum concentration was not considered statistically positive at the 3s level),
- $^{60}\text{Co}$ ,  $^{241}\text{Am}$ ,  $^{238}\text{Pu}$ , and  $^{239,240}\text{Pu}$  were present in Interval 2 at concentrations statistically larger than background, and
- anthropogenic radionuclide concentrations in Interval 3 were not statistically larger than those in background wells.

Another approach, described only briefly by Dames and Moore (1992, p. 74) provides additional, qualitative evidence of migration of radionuclides at the SDA. "The evidence supporting the conclusion of contaminant transport from the SDA was investigated qualitatively by conducting a between-interval comparison for the radionuclides identified above. The approach was similar to the qualitative assessment of repeatability between intervals, but was applied vertically. If a positive result for a specific radionuclide is observed between intervals at a specific location, this was considered supporting evidence of contaminant migration. Thus, the results from this evaluation provide qualitative evidence that radionuclides have been transported from the buried waste at the SDA. The following radionuclides can be correlated vertically at the SDA for the locations indicated: (a)  $^{241}\text{Am}$  was detected at well USGS 93 in Intervals 1 and 2; (b)  $^{241}\text{Am}$  was detected in well D-02 in Interval 1 and in Interval 2 at both wells 79-2 and TW-1 (note: wells D-02, 79-1, and TW-1 were grouped for comparison because of their close proximity

to each other); (c)  $^{239,240}\text{Pu}$  was observed in well D-02 in Interval 1 and wells 79-2 and TW-1 in Interval 2; (d)  $^{239,240}\text{Pu}$  was observed in well D-02 in Interval 3; and (e)  $^{238}\text{Pu}$  was observed in Interval 1 at well D-02, in Interval 2 at well TW-1, and in Interval 3 at well D-02."

It should be noted here that the criterion used for these detections (at 3s level) is less rigorous than the repeatable, statistically positive (3s) criterion used earlier in the Dames and Moore (1992, p. 66) report. For example, although the results listed above invoke Interval 1 data, there were no repeatable, statistically positive results in this interval.

(9) Additional investigations—Late in our review process, we learned of additional data. Becker and others (1996, p. 4–78) noted a 1993 study (Loehr and others, 1993) of archived cores collected from the SDA during 1971–89. Time did not permit detailed reanalysis of the data from that study. These data, originally were analyzed by a contract laboratory (Barringer Laboratories, Denver, Colorado), but were not included in the 1992–94 Dames and Moore compilation. Fifty-four samples from 21 wells were analyzed for radionuclides, including  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$ . Samples came from massive, unfractured basalts, basalt rubble zones, sediment-filled fractured basalt, and sedimentary interbed material from the A-B, B-C, and C-D interbeds. The data do not contain counting error statistics; these apparently were not provided by the laboratory. In 4 to 25 percent of the samples from the fractured basalts, rubble zones, and sedimentary interbeds, the concentrations of  $^{241}\text{Am}$  exceeded the stated detection limit (0.12 pCi/g). These were the only anthropogenic actinide detections (uranium isotopes excluded), and their statistical significance cannot be evaluated because of the omission of counting error data.

#### 3.6.2 Perched-water and ground-water data

Perched water has been predominately located and sampled in 6 of the 45 wells completed within the RWMC (Scott Barrie, written commun., 1998). These perched water zones are primarily above the C-D interbed, and five wells routinely yield sufficient volumes of water for analyses; these wells are USGS 92, 88-02D, D-10, 78-1 and 77-2 (figs. 2-2 and 2-3). In addition to these wells, about 25 lysimeters are located within the SDA and are sampled for radionuclides. All but one of the lysimeters that yield water are within the perimeter of the SDA and are completed to depths of from about 2 to 7 m. The one lysimeter not within the SDA that yields water is about 30 m outside the perimeter. Since 1972, the site contractor at the INEEL has sampled water from these perched wells

and lysimeters at the RWMC. In that time, a total of 14 different radionuclides, including transuranics, have been detected at least once. These detections were based on the use of the 2s detection criteria as outlined in section 3.2 titled "Detection limits, statistical screening criteria, and reporting of data." Reported detections of selected radionuclides since 1972 are summarized in table 3-14.

Table 3-14. Number of radionuclide detections in water samples from the perched water zone at the Radioactive Waste Management Complex using the site contractor's criterion of detection, 2s and greater (Scott Barrie, written commun., 1998)

Radionuclide	Total number of samples	Number of detections
$^{241}\text{Am}$	75	5
$^{233,234}\text{U}$	16	7
$^{235}\text{U}$	46	7
$^{238}\text{U}$	19	7
$^{238}\text{Pu}$	43	8
$^{239,240}\text{Pu}$	40	8
$^{230}\text{Th}$	8	3

These data were provided by LMITCO and were reviewed first with the criteria of detection used by them (2s) and then using the method recommended by the National Institute of Standards Technology (NIST), ASTM, and IUPAC (3s and greater after reviewing all data between 1.6s and 3s). With so few detections reported for a given radionuclide, a rigorous statistical analysis was not possible, but some general observations can be made.

Three of the five (table 3-14) reported detections for  $^{241}\text{Am}$  were in water or sediments from well USGS 92 (fig. 2-3). This well is completed to a depth of 65.5 m and is within the SDA. The reported detections in water from USGS 92 were  $0.00041 \pm 0.00012$  pCi/L in 1976 and  $0.07 \pm 0.03$  pCi/L in 1981; the reported detection in sediment was  $0.0312 \pm 0.0129$  pCi/kg in 1998. There is a transcription problem with the 1976 value, which was correctly reported by the USGS as  $0.041 \pm 0.012$  pCi/L. It is not correct to compare this value to the method detection limit used by LMITCO (0.05 pCi/L). By hypothesis testing this value may be a detection. Additionally, in 1997, there was a reported detection from well W-25 of  $0.629 \pm 0.233$  pCi/L. The remaining detection of  $^{241}\text{Am}$  reported for water from the perched zone at the SDA was from lysimeter PA-03; the completion depth of this lysim-

eter is 3 m. The  $^{241}\text{Am}$  concentration in water from PA-03 in 1998 was  $4.16 \pm 0.656$  pCi/L. This concentration is a valid detection by the application of hypothesis testing.

Six of the seven reported detections of  $^{233,234}\text{U}$  were also in water from well D-06 and lysimeter PA-03. The concentrations of  $^{233,234}\text{U}$  in the six water samples collected from D-06 and PA-03 in 1997–98 ranged from  $8.45 \pm 1.27$  to  $84.4 \pm 6.23$  pCi/L. The remaining reported detection of  $^{233,234}\text{U}$  ( $90 \pm 7.29$  pCi/L) was in water collected from well TW-1 at a depth of 102 ft in February 1998.

Seven detections of  $^{235}\text{U}$  were reported. The reported detections for  $^{235}\text{U}$  were in water samples from two perched-water wells (D-06 and TW-1) and two lysimeters (PA-03 and W-08). The  $^{235}\text{U}$  concentrations ranged from  $1.61 \pm 0.377$  pCi/L in water from lysimeter PA-03 to  $62.2 \pm 25.5$  pCi/L in water from lysimeter W-08. Seven detections of  $^{238}\text{U}$  were also reported for water collected from the same two wells (D-06 and TW-1) and the two lysimeters (PA-03 and W-08) as the  $^{235}\text{U}$  detections. The  $^{238}\text{U}$  concentrations ranged from  $7.2 \pm 1.15$  to  $49.4 \pm 3.75$  pCi/L.

Eight detections of  $^{238}\text{Pu}$  were reported. Because the reported inventories of  $^{238}\text{Pu}$  at the SDA are about two orders of magnitude smaller than inventories reported for  $^{239,240}\text{Pu}$  (Becker and others, 1998, table 4-1), any detection of  $^{238}\text{Pu}$  in environmental samples at the SDA is questionable, especially in samples without detections of  $^{239,240}\text{Pu}$ . The reported detections for  $^{238}\text{Pu}$  were in seven water samples and one sediment sample from one lysimeter (PA-03) and three wells (USGS 92, D-06, and D-15; the completion depth for D-06 is 26.8 m, and for D-15 is 29.9 m). The  $^{238}\text{Pu}$  concentrations ranged from  $0.0063 \pm 0.0006$  pCi/L in 1977 in water from USGS 92 to  $24 \pm 2.05$  pCi/L in water from PA-03 in 1997. The concentration of  $^{238}\text{Pu}$  in the sediment sample was reported as  $1.39 \pm 0.285$  pCi/kg.

There were also eight reported detections of  $^{239,240}\text{Pu}$  in water from wells and lysimeters in the perched-water zone at the SDA. Three of the detections were in sediment samples and three were in water samples collected from USGS 92. The remaining two detections were in water samples from lysimeter PA-03 and well D-15. The concentrations ranged from  $0.023 \pm 0.008$  pCi/L to  $1.06 \pm 0.351$  pCi/L. Because the stated method detection limit was 0.05 pCi/L, three of the reported detections for  $^{239,240}\text{Pu}$  probably were not true detections. Concentrations in all three of the water samples collected at USGS 92 were smaller than the method detection limit and, therefore, should be viewed as nondetections. Information on recovery and counting times for analysis of these samples was

### 3-12 Review of the transport of selected radionuclides in the Interim Risk Assessment

unavailable. For these reasons these three results must be considered as nondetections.

The three detections reported for  $^{230}\text{Th}$  were in water collected from one perched-water well (D-06) and one lysimeter (PA-03). In August 1997, water collected from the well had a  $^{230}\text{Th}$  concentration of  $1.18 \pm 0.477$  pCi/L. Water collected from lysimeter PA-03 in August 1997 had a  $^{230}\text{Th}$  concentration of  $0.857 \pm 0.367$  pCi/L; additionally, water collected from PA-03 in February 1998 had a concentration of  $0.943 \pm 0.394$  pCi/L.

These same data were reviewed using the recommended method outlined previously. The number of detections reported by the site contractor for  $^{241}\text{Am}$  and  $^{238}\text{Pu}$  in water and sediment samples from the unsaturated zone remained the same. The number of detections for  $^{239,240}\text{Pu}$  decreased to three and the largest concentration was then 0.7 pCi/L. Regardless of the detection criteria applied to these data, too few results were reported to definitively and statistically confirm the occurrence of radionuclides in water and sediments from wells and lysimeters at the SDA. However, it is not prudent or scientifically defensible to ignore true statistical detections. It is recommended that samples be collected during episodic recharge events and that sampling frequency be increased in areas where detections periodically have been reported. All of the reported  $^{233,234}\text{U}$ , all but two of the reported  $^{235}\text{U}$ , and all of the reported  $^{238}\text{U}$  concentrations were still detections. None of the reported  $^{230}\text{Th}$  concentrations were detections.

In 1992, the site contractor completed six groundwater monitoring wells in the Snake River Plain aquifer at the SDA. Between October 1992 and March 1998 two detections for  $^{239,240}\text{Pu}$  and five detections for  $^{241}\text{Am}$  were reported. The detections for  $^{239,240}\text{Pu}$  were  $1.3 \pm 0.3$  and  $4.3 \pm 0.5$  pCi/L. The concentration of  $1.3 \pm 0.3$  pCi/L is questionable because the associated uncertainty is almost 30 percent of the result, and the result is about 1.5 orders of magnitude larger than the method detection limit. The associated uncertainty should not be this large. The same concentration was repeated with an  $^{241}\text{Am}$  result that was discussed in section 3.2 titled "Detection limits, statistical screening criteria, and reporting of data." The  $^{241}\text{Am}$  concentrations ranged from  $0.045 \pm 0.013$  to  $1.13 \pm 0.3$  pCi/L. The smallest concentration,  $0.045 \pm 0.013$  pCi/L, is questionable because such precision is not possible for a concentration that is less than the method detection limit. For apparent statistical detections at and below the method detection limit, more QA/QC information should be provided for a defensible review.

### 3.6.3 Significance of reported detections

Regardless of the criteria used to report detections, there have been positive occurrences of radionuclides in the environment at the SDA. The source of the radionuclides is difficult to establish because of the small number of statistically positive results. Possible sources include cross contamination during sampling, radionuclides carried to the subsurface during well completion, laboratory contamination, global fallout from weapons tests of the 1950–60's, true radionuclide migration from buried waste at the SDA, and wastes injected into the aquifer upgradient from the SDA at the TRA or INTEC. One way to help delineate the source(s) of detected radionuclides would be to determine isotopic ratios of plutonium and americium in the environment. To better understand these apparent detections in the subsurface at the SDA, increased sampling, lower method detection limits, and increased cooperation between researchers is needed.

## 3.7 Summary

The following recommendations are offered in an attempt to improve sample collection and analyses, data generation and reporting, and overall quality assurance and quality control for the program that has been charged with generating data in support of the WAG-7 Comprehensive RI/FS.

(1) The decision as to whether to use a 2s or 3s detection (with consideration of concentrations between 1.6 and 3s) criterion has apparently yet to be resolved at the INEEL (see Becker and others (1996; p. 4-76 to 4-77)). To meet the requirements of the National Technology Transfer and Advancement Act (Public Law 104-113), the method recommended by ASTM should be considered and applied to radionuclide data sets as a minimum screening criteria (see section 3.4 of this report). Additionally, warning flags for exotic radionuclide isotopes should be included in the computer data base and in reports to the DOE and the State of Idaho.

(2) Data archiving problems were identified. Compilations of data, which tend to supplant original data, need greater QA. Additionally, all data generated in support of this program should be made available to the INEEL Earth-science community for review at the earliest time possible after completion and a thorough QA/QC check.

(3)  $^{237}\text{Np}$  was highlighted in the SOW as a radionuclide of concern. This concern presumably stems from the long half life ( $2.1 \times 10^6$  years), high radiotoxicity, significant inventory in wastes, and the solubility and mobility of  $\text{Np}^{5+}$  species in natural waters (Langmuir, 1997;

Bertetti and others, 1998); however, no data for  $^{237}\text{Np}$ , in either ground water or interbed sediments at the SDA was presented for review. If monitoring for  $^{237}\text{Np}$  is not currently a part of this monitoring program, then it should be added.

(4) Thermal ionization mass spectrometry and (or) accelerator mass spectrometry for the determination of Pu isotopes in ground water and sediment samples may allow for lower detection limits than those achievable with  $\alpha$ -spectroscopy. Lower detection limits could be of value in future sampling of the water and sediments at WAG-7. Use of Pu isotope ratios may help to resolve source issues (such as waste versus global fallout) at sites like well

USGS 87, which are upgradient and outside the SDA perimeter.

(5) A consistent method of data validation needs to be implemented. To date, all levels of validation from A through C have been applied to radionuclide data.

(6) The reporting of more precision in the uncertainty of an analytical result than is reported for the analytical result should be avoided. For example, a  $^{239,240}\text{Pu}$  concentration of  $1.06 \pm 0.351$  pCi/L was reported for perched water at the RWMC. It would be more appropriate to report the uncertainty of the measurement as 0.35 or 0.4 pCi/L.

### 3-14 Review of the transport of selected radionuclides in the Interim Risk Assessment

Table 3-1. Concentrations of selected radionuclides in sediment samples collected in 1972 from wells at and near the Subsurface Disposal Area (from Barraclough and others, 1976; table 2)

[Analyses performed at the Radiological and Environmental Sciences Laboratory. Sample depth intervals are in feet (ft) and inches (in.); concentrations are in microcuries per gram times  $10^{-9}$ . Symbols: <, less than;  $\pm$ , plus or minus; [a] = rerun on different aliquot; [b] = ND, not detected; [c] = NA, not analyzed; and [d] = statistically positive at the 95 percent confidence level]

Well	Sample depth interval	$^{60}\text{Co}$	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{140}\text{Ba/La}$	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$	$^{241}\text{Am}$
USGS 87	231 ft 2 in.-233 ft	<50	460 $\pm$ 50 [d]	<50	[b]	<4.0	29.0 $\pm$ 4.5 [d]	<2.0
USGS 88	521 ft-522 ft	[b]	400 $\pm$ 90 [d]	[b]	[b]	15.0 $\pm$ 3.0 [d]	22.0 $\pm$ 3.0 [d]	5.0 $\pm$ 3.0
USGS 89	241 ft 7 in.-243 ft 2 in.	<3	640 $\pm$ 70 [d]	40 $\pm$ 10 [d]	[b]	[c]	<1	2.0 $\pm$ 3.0
	540 ft-545 ft	[b]	300 $\pm$ 90 [d]	[b]	[b]	[b]	[b]	[b]
USGS 91	7 ft 10 in.-8 ft 11 in.	[b]	<4,200	[b]	[b]	[c]	26.0 $\pm$ 4.5 [d]	[c]
	233 ft 9 in.-236 ft 3 in.	<30	1,200 $\pm$ 100 [d]	<30	[b]	3.0 $\pm$ 1.5	0.5 $\pm$ 1.0	12.0 $\pm$ 4.5 [d]
	236 ft 6 in.-237 ft	[b]	<3,300	[b]	[b]	[c]	140.0 $\pm$ 7.0 [d]	[c]
	243 ft 2 in.-245 ft 1 in.	<90	500 $\pm$ 90 [d]	<200	[b]	9.0 $\pm$ 7.0	2.0 $\pm$ 3.0	5.0 $\pm$ 3.0
USGS 92	5 ft-7 ft 6 in.	[b]	240 $\pm$ 70 [d]	[b]	[b]	[c]	<1.0	[c]
	88 ft 6 in.-90 ft	<20	300 $\pm$ 90 [d]	<20	[b]	6.0 $\pm$ 3.0	0.5 $\pm$ 1.5	1.0 $\pm$ 1.5
	223 ft-225 ft 6 in.	230 $\pm$ 20 [d]	300 $\pm$ 90 [d]	130 $\pm$ 30 [d]	[b]	3.0 $\pm$ 3.0	7.0 $\pm$ 3.0 [d]	2.0 $\pm$ 1.5
USGS 93	13 ft 10 in.-14 ft	<60	400 $\pm$ 90 [d]	<40	48 $\pm$ 12 [d]	0.2 $\pm$ 0.3	1.0 $\pm$ 1.5	12.0 $\pm$ 4.5 [d]
	98 ft-101 ft	62 $\pm$ 12 [d]	<300	100 $\pm$ 20 [d]	[b]	8.0 $\pm$ 1.5 [d]	110.0 $\pm$ 7.0 [d]	[c]
	101 ft-103 ft	<30	690 $\pm$ 110 [d]	<50	[b]	9.0 $\pm$ 3.0 [d]	230.0 $\pm$ 11.0 [d]	63.0 $\pm$ 12.0 [d]
	101 ft-103 ft [a]	[b]	<200	[b]	[b]	14.0 $\pm$ 3.0 [d]	540.0 $\pm$ 12.0 [d]	150.0 $\pm$ 22.0 [d]
	103 ft-105 ft	<310	400 $\pm$ 100 [d]	<900	[b]	[c]	81.0 $\pm$ 11.0 [d]	45.0 $\pm$ 6.0 [d]
USGS 94	98 ft 4 in.-98 ft 5 in.	[b]	150 $\pm$ 50 [d]	[b]	[b]	[b]	[b]	[b]
	262 ft 3 in.-264 ft 7 in.	250 $\pm$ 30 [d]	<200	180 $\pm$ 30 [d]	[b]	4.0 $\pm$ 3.0	3.0 $\pm$ 1.5	1.0 $\pm$ 1.5
USGS 95	20 ft-22 ft	<50	200 $\pm$ 90 [d]	<40	[b]	5.0 $\pm$ 3.0	3.0 $\pm$ 1.5	2.0 $\pm$ 3.0
	112 ft-113 ft 4 in.	<30	<200	220 $\pm$ 10 [d]	[b]	0.5 $\pm$ 1.0	0.5 $\pm$ 1.0	3.0 $\pm$ 4.5
	226 ft 9 in.-229 ft 3 in.	240 $\pm$ 30 [d]	<200	230 $\pm$ 30 [d]	[b]	9.0 $\pm$ 7.0	9.0 $\pm$ 7.0	3.0 $\pm$ 3.0
USGS 96	100 ft 6 in.-101 ft	<20	<200	<40	[b]	6.0 $\pm$ 1.5 [d]	45.0 $\pm$ 2.0 [d]	10.0 $\pm$ 7.0
	110 ft-112 ft 11 in.	<20	<200	<30	[b]	3.0 $\pm$ 1.5	3.0 $\pm$ 1.5	230.0 $\pm$ 20.0 [d]
	110 ft-110 ft 6 in.	<70	---	<40	[b]	9.0 $\pm$ 7.0	0.5 $\pm$ 1.0	30.0 $\pm$ 6.0 [d]
	122 ft 9 in.-123 ft 10 in.	<70	200 $\pm$ 100	550 $\pm$ 40 [d]	[b]	2.0 $\pm$ 1.5	2.0 $\pm$ 1.5	5.0 $\pm$ 3.0
	124 ft-124 ft 3 in.	<90	200 $\pm$ 90 [d]	<90	[b]	[c]	<0.5	3.0 $\pm$ 4.5
	221 ft 7 in.-224 ft 1 in.	<30	200 $\pm$ 100	<40	[b]	1.0 $\pm$ 3.0	4.0 $\pm$ 1.5 [d]	6.0 $\pm$ 3.0

Table 3-2. Concentrations of selected radionuclides in sediment samples collected in 1975 from wells USGS 93 and USGS 96 (from Burgus and Maestas, 1976; table 4)

[Analyses performed at the Radiological and Environmental Sciences Laboratory. Sample depths are in feet. Concentrations are in microcuries per gram times  $10^{-9}$  ( $10^{-9}$   $\mu\text{Ci/g}$ ) for  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$  and in microcuries per gram times  $10^{-8}$  ( $10^{-8}$   $\mu\text{Ci/g}$ ) for  $^{90}\text{Sr}$ ,  $^{144}\text{Ce}$ ,  $^{137}\text{Cs}$ , and  $^{60}\text{Co}$ ]

Sample no.	Well	Sample depth	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$	$^{241}\text{Am}$	$^{90}\text{Sr}$	$^{144}\text{Ce}$	$^{137}\text{Cs}$	$^{60}\text{Co}$
1	96A	34.8-35.1	0.6 $\pm$ 1.1	0.1 $\pm$ 0.6	-0.6 $\pm$ 1.8	-10 $\pm$ 4	16 $\pm$ 11	-5 $\pm$ 3	2 $\pm$ 2
2	96B	35-40	0.5 $\pm$ 1.1	0.2 $\pm$ 0.6	-0.9 $\pm$ 1.6	0 $\pm$ 5	0 $\pm$ 7	0 $\pm$ 1	-0.3 $\pm$ 1.0
3	96B	36.0	-0.5 $\pm$ 1.0	0.1 $\pm$ 0.6	-1.5 $\pm$ 1.4	-6 $\pm$ 5	0 $\pm$ 10	-1 $\pm$ 3	-3 $\pm$ 2
4	96B	101.6-102.0	0.0 $\pm$ 1.1	0.5 $\pm$ 0.6	-1.2 $\pm$ 1.5	-8 $\pm$ 5	10 $\pm$ 10	-2 $\pm$ 3	-3 $\pm$ 2
5	96B	106.4-106.8	0.7 $\pm$ 1.1	0.1 $\pm$ 0.6	-0.7 $\pm$ 1.6	-6 $\pm$ 5	20 $\pm$ 10	1 $\pm$ 2	2 $\pm$ 2
6	96B	220.9-221.2	0.4 $\pm$ 1.4	0.1 $\pm$ 0.6	0.6 $\pm$ 1.5	-2 $\pm$ 5	10 $\pm$ 10	1 $\pm$ 2	2 $\pm$ 2
7	96B	223.6-223.9	0.6 $\pm$ 1.1	0.7 $\pm$ 0.7	1.5 $\pm$ 1.8	3 $\pm$ 5	6 $\pm$ 9	2 $\pm$ 2	2 $\pm$ 2
8	93A	80.2-80.6	0.5 $\pm$ 1.5	-0.3 $\pm$ 0.5	0.4 $\pm$ 1.6	-2 $\pm$ 5	10 $\pm$ 10	4 $\pm$ 2	2 $\pm$ 2
9	93A	82.8-83.6	0.2 $\pm$ 1.0	1.0 $\pm$ 0.8	-0.3 $\pm$ 1.6	-2 $\pm$ 5	20 $\pm$ 10	2 $\pm$ 2	1 $\pm$ 3
10	93A	89.0-89.5	-0.4 $\pm$ 1.0	-0.3 $\pm$ 0.5	-0.8 $\pm$ 1.6	7 $\pm$ 5	4 $\pm$ 13	-1 $\pm$ 2	2 $\pm$ 2
11	93A	97.9-98.0	2.0 $\pm$ 1.6	0.0 $\pm$ 0.6	0.5 $\pm$ 1.7	2 $\pm$ 4	10 $\pm$ 10	1 $\pm$ 2	5 $\pm$ 3
12	93A	100.6-100.9	2.0 $\pm$ 1.5	0.0 $\pm$ 0.6	-0.8 $\pm$ 1.6	2 $\pm$ 8	10 $\pm$ 10	-1 $\pm$ 2	1 $\pm$ 2
13	93A	102.4-102.8	0.8 $\pm$ 1.2	0.4 $\pm$ 0.6	-1.6 $\pm$ 1.6	6 $\pm$ 4	10 $\pm$ 10	-2 $\pm$ 2	1 $\pm$ 2
14	93A	221.5-223.5	-0.4 $\pm$ 1.0	0.1 $\pm$ 0.5	-0.6 $\pm$ 1.6	5 $\pm$ 5	-10 $\pm$ 10	2 $\pm$ 2	2 $\pm$ 2
15	93A	226.5-226.9	-0.3 $\pm$ 1.0	0.3 $\pm$ 0.5	0.1 $\pm$ 1.6	7 $\pm$ 4	-10 $\pm$ 10	-2 $\pm$ 3	-5 $\pm$ 2



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**Table 3-3. Concentrations of selected radionuclides in sediment samples collected in 1976 from wells 76-1, 76-2, and 76-3 (from Humphrey and Tingey, 1978; Table 1)**

[Analyzed by Allied Chemical Corporation. Sample depths are in feet. Concentrations are in microcuries per gram times  $10^{-9}$  ( $10^{-9}$   $\mu\text{Ci/g}$ ) for  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$  and in microcuries per gram times  $10^{-8}$  ( $10^{-8}$   $\mu\text{Ci/g}$ ) for  $^{90}\text{Sr}$ ,  $^{144}\text{Ce}$ ,  $^{137}\text{Cs}$ , and  $^{60}\text{Co}$ . Symbols: [a] = sample not analyzed; [b] = statistically positive at the 95 percent confidence level; [c] = sample material not available for rerun; and [d] = sample reanalyzed (see table 3-5)]

Sample no.	Sample depth	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$	$^{241}\text{Am}$	$^{90}\text{Sr}$	$^{144}\text{Ce}$	$^{137}\text{Cs}$	$^{60}\text{Co}$
76-1-1	35.1	0.3±15.8	-10.8±20.1	[a]	5±4	5±15	10±6	2±4
76-1-2	29.2	-2.4±15.6	-29.2±19.9	[a]	-2±3	-1±13	7±8	-0.1±3
76-1-3	84.7-86.3	2.3±4.8	6.8±5.3	[a]	-6±5	60±111	0±4	0±7
76-1-4	92.3-93.7	1.7±5.5	2.2±5.9	[a]	-3±5	0±20	-7±3	2±4
76-1-5	92.3	2.07±16.4	13.2±6.1 [b,c]	[a]	[a]	0±50	0±9	11±11
76-1-6	103.0-104.5	9.9±7.1	4.5±6.6	[a]	-10±5	0±20	-1±5	4±4
76-1-7	217.2	10.7±4.2 [b,d]	-4.8±4.0	[a]	-5±3	10±20	0±4	0±2
76-1-8	221.0	6.3±16.4	14.4±21	[a]	49±6 [b,d]	0±10	1±2	-2±2
76-1-9	228.3	3.3±15.4	-1.7±20	2±1.6	7±8	2±11	-5±4	4±3
76-1-10	70.0-75.0	13.0±15.8	-21.6±20	[a]	[a]	0±7	-3±2	0±3
76-2-1	16.0	5.7±5.6	-7.1±5.7	[a]	-7±5	3±12	-3±3	-2±3
76-2-2	24.0	7.3±6.9	-10.7±6.3	[a]	-4±5	20±20	5±4	0±4
76-2-3	24.3	33.0±16.6	1.1±5.4	[a]	1±4	-6±14	-4±2	-1±2
76-2-4	60.0	12.7±4.6 [b,d]	-2.9±4.3	[a]	[a]	-5±12	8±3 [b,d]	2±4
76-2-5	82.5-83.5	5.7±4.0	11.5±4.4 [b,d]	[a]	-4±4	11±15	-7±3	0±4
76-2-6	223.0	5.9±15.6	1.7±20.1	[a]	3±4	0±10	5±3	1±2
76-2-7	235.0	1.8±15.7	0.7±20.1	[a]	8±6	-0.3±14	-5±5	-3±4
76-2-8	243.9	-0.6±15.5	6.6±19.9	[a]	-2±6	-7±16	2±6	6±4
76-2-9	245.8	13.2±15.7	23.5±20.1	[a]	-7±6	-9±9	-2±3	2±2
76-3-1	24.0-24.7	0.0±1.1	-0.6±1.0	0.0±1.2	0.4±4	-5±11	-7±4	1±3
76-3-2	25.8-26.8	-0.1±1.2	0.2±1.0	1±2	5±4	18±12	-8±5	6±3
76-3-3	96.5-96.8	-0.1±1.2	-0.7±1.1	0.3±1.4	9±4 [b,d]	0±10	-5±3	0±3
76-3-4	97.5-97.8	8.7±4.4	16.8±5.0 [b,d]	8.4±1.4 [b,d]	7±4	0±10	6±3	0±2
76-3-5	110.5-111.0	1.0±1.2	-0.4±1.1	2.7±1.2 [b,c]	4±3	23±13	1±5	2±3
76-3-6	119.5-119.7	26.3±17	2.0±20.9	[a]	-7±4	2±3	4±4	6±4
76-3-7	222.5	23.7±15.8	-8.4±20	[a]	20±8 [b,d]	16±14	3±3	3±3
76-3-8	228.5	19.3±15.7	-7.4±19.9	[a]	5±4	40±20	1±3	2±3
76-3-9	240.4	1.3±4.5	10.7±5.6	[a]	7±4	3±10	2±3	9±5

Table 3-4. Concentrations of selected radionuclides in sediment samples collected in 1976 from wells 76-4, 76-4A, 76-5, 76-6, and 77-2 (from Humphrey and Tingey, 1978; table 2)

[Analysis performed at the Radiological and Environmental Sciences Laboratory. Sample depths are in feet. Concentrations are in microcuries per gram times  $10^{-9}$  ( $10^{-9}$   $\mu\text{Ci/g}$ ) for  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$  and in microcuries per gram times  $10^{-8}$  ( $10^{-8}$   $\mu\text{Ci/g}$ ) for  $^{90}\text{Sr}$ ,  $^{144}\text{Ce}$ ,  $^{137}\text{Cs}$ , and  $^{60}\text{Co}$ . Symbols: [a] = sample not analyzed, and [b] = statistically positive at the 95 percent confidence level (see table 3-5 for reanalysis)]

Sample no.	Sample depth	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$	$^{241}\text{Am}$	$^{90}\text{Sr}$	$^{144}\text{Ce}$	$^{137}\text{Cs}$	$^{60}\text{Co}$
76-4-1	12.0	0.1 $\pm$ 0.9	1.3 $\pm$ 0.9	0.0 $\pm$ 1.0	1 $\pm$ 3	-100 $\pm$ 100	1 $\pm$ 2	2 $\pm$ 2
76-4-2	20.0	0.0 $\pm$ 0.8	0.4 $\pm$ 0.6	-0.2 $\pm$ 0.9	2 $\pm$ 3	5 $\pm$ 9	1 $\pm$ 2	1 $\pm$ 2
76-4-3	99.5	0.5 $\pm$ 0.9	3.1 $\pm$ 1.3 [b]	0.5 $\pm$ 0.8	1 $\pm$ 3	10 $\pm$ 20	4 $\pm$ 2	2 $\pm$ 2
76-4A-1	23.5	0.0 $\pm$ 0.7	1.0 $\pm$ 1.0	21 $\pm$ 3 [b]	-4 $\pm$ 3	-1 $\pm$ 2	-2 $\pm$ 10	0 $\pm$ 2
76-4A-2	97.8	1.3 $\pm$ 1.0	0.5 $\pm$ 0.5	6.4 $\pm$ 1.7 [b]	-4 $\pm$ 3	20 $\pm$ 20	8 $\pm$ 4	0 $\pm$ 2
76-4A-3	223.5	0.7 $\pm$ 0.9	1.2 $\pm$ 0.8	0.0 $\pm$ 1.0	0 $\pm$ 3	-14 $\pm$ 10	0 $\pm$ 3	1 $\pm$ 2
76-4A-4	226.0	1.0 $\pm$ 1.0	-0.2 $\pm$ 0.5	23 $\pm$ 3 [b]	1 $\pm$ 3	40 $\pm$ 30	2 $\pm$ 3	2 $\pm$ 2
76-5-1	25.4-26.0	2.5 $\pm$ 1.5	0.0 $\pm$ 1.0	0.6 $\pm$ 1.0	0 $\pm$ 3	30 $\pm$ 20	2 $\pm$ 2	2 $\pm$ 2
76-5-2	75.1	2.0 $\pm$ 2.0	2.0 $\pm$ 3.0	-1.0 $\pm$ 1.0	0 $\pm$ 3	-10 $\pm$ 20	2 $\pm$ 2	2 $\pm$ 2
76-5-3	95.9	2.0 $\pm$ 2.0	0.0 $\pm$ 1.0	1.0 $\pm$ 1.0	2 $\pm$ 3	2 $\pm$ 9	1 $\pm$ 2	2 $\pm$ 2
76-5-4	223.7	0.8 $\pm$ 0.7	-0.7 $\pm$ 0.5	-1.0 $\pm$ 1.0	-2 $\pm$ 6	8 $\pm$ 10	2 $\pm$ 2	2 $\pm$ 2
76-5-5	114.3-114.8	0.6 $\pm$ 0.9	0.8 $\pm$ 0.7	0.0 $\pm$ 1.0	3 $\pm$ 3	4 $\pm$ 9	-1 $\pm$ 2	1 $\pm$ 2
76-6-1	17.1	0.0 $\pm$ 1.0	0.1 $\pm$ 0.6	-0.9 $\pm$ 0.7	4 $\pm$ 4	-15 $\pm$ 9	1 $\pm$ 2	3 $\pm$ 2
76-6-2	102.1	3.0 $\pm$ 2.0	1.4 $\pm$ 1.1	-1.4 $\pm$ 1.5	9 $\pm$ 5	-10 $\pm$ 20	3 $\pm$ 2	-2 $\pm$ 2
76-6-3	232.2-233.2	2.9 $\pm$ 1.4 [b]	0.9 $\pm$ 0.9	0.0 $\pm$ 2.0	-3 $\pm$ 5	-25 $\pm$ 13	-2 $\pm$ 3	4 $\pm$ 3
76-6-4	236.3	-0.2 $\pm$ 0.8	0.5 $\pm$ 0.7	0.0 $\pm$ 2.0	2 $\pm$ 3	10 $\pm$ 10	1 $\pm$ 2	1 $\pm$ 2
77-2-1	19.2	3.0 $\pm$ 2.0	0.0 $\pm$ 2.0	2.0 $\pm$ 2.0	[a]	60 $\pm$ 30	2 $\pm$ 6	4 $\pm$ 4
77-2-2	27.7	-0.4 $\pm$ 0.7	0.9 $\pm$ 0.8	1.9 $\pm$ 1.3	8 $\pm$ 4	10 $\pm$ 10	1 $\pm$ 2	2 $\pm$ 2
77-2-3	25.8-27.7	0.6 $\pm$ 0.9	1.5 $\pm$ 0.9	-0.7 $\pm$ 0.7	5 $\pm$ 4	-9 $\pm$ 14	7 $\pm$ 3 [b]	0 $\pm$ 2

### 3-18 Review of the transport of selected radionuclides in the Interim Risk Assessment

Table 3-5. Concentrations of radionuclides  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{90}\text{Sr}$ , and  $^{137}\text{Cs}$  in split samples reanalyzed on the basis of initially statistically positive results listed in tables 3-3 and 3-4 (from Humphrey and Tingey, 1978; table 5)

[Analysis performed at the Radiological and Environmental Sciences Laboratory. Sample depth is in feet. Concentrations are in microcuries per gram times  $10^{-9}$  ( $10^{-9}$   $\mu\text{Ci/g}$ ) for  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$  and in microcuries per gram times  $10^{-8}$  ( $10^{-8}$   $\mu\text{Ci/g}$ ) for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . Abbreviations: [a] = statistically positive at 95% confidence level but not at the 99% confidence level, and [b] = statistically positive at the 99% confidence level]

Sample no.	Sample depth	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$	$^{241}\text{Am}$	$^{90}\text{Sr}$	$^{137}\text{Cs}$
76-1-7	217.3	10.7 $\pm$ 4.2 [a] 0.0 $\pm$ 1.0				
76-1-8	221.2				49 $\pm$ 6 [b] 42 $\pm$ 5 [b]	
76-2-4	60.0	12.7 $\pm$ 4.6 [b] 0.0 $\pm$ 2.0				8 $\pm$ 3 [a] 10 $\pm$ 10
76-2-5	82.5-83.5		11.5 $\pm$ 4.4 [a] 0.0 $\pm$ 1.0			
76-3-3	96.5-96.8				9 $\pm$ 4 [a] 1 $\pm$ 3	
76-3-4	97.5-97.8		16.8 $\pm$ 5.0 [b] 0.0 $\pm$ 1.0	8.4 $\pm$ 1.4 [b] -1.0 $\pm$ 1.0		
76-3-7	222.5				20 $\pm$ 8 [a] 2 $\pm$ 4	
76-4-3	99.5		3.1 $\pm$ 1.3 [a] -0.7 $\pm$ 0.9			
76-4A-1	23.5			21 $\pm$ 3 [b] 0.0 $\pm$ 1.0 -1.0 $\pm$ 1.0		
76-4A-2	97.8			6.4 $\pm$ 1.7 [b] -0.0 $\pm$ 1.0 0.0 $\pm$ 1.0		
76-4A-4	226.0			23 $\pm$ 3 [b] 2.3 $\pm$ 1.5 -1.0 $\pm$ 1.0		
76-6-3	232.2-233.2	2.9 $\pm$ 1.4 [a] 0.0 $\pm$ 1.0				
77-2-3						7 $\pm$ 3 [a] -5 $\pm$ 3

Table 3-6. Concentrations of radionuclides  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{90}\text{Sr}$ ,  $^{144}\text{Ce}$ ,  $^{137}\text{Cs}$ , and  $^{60}\text{Co}$  in sediment samples collected in 1978 from wells 78-2, 78-3, and 78-5 (from Humphrey, 1980; Table 1)

[Analysis performed at the Radiological and Environmental Sciences Laboratory. Sample depths are in meters. Concentrations are in microcuries per gram times  $10^{-8}$  ( $10^{-8}$   $\mu\text{Ci/g}$ ) for  $^{90}\text{Sr}$ ,  $^{144}\text{Ce}$ ,  $^{137}\text{Cs}$ , and  $^{60}\text{Co}$  and in microcuries per gram times  $10^{-9}$  ( $10^{-9}$   $\mu\text{Ci/g}$ ) for  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$ . Abbreviation: [a] = statistically positive at the 95 percent confidence level; (A,B) = splits of the original sample]

Sample no.		Sample depth	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$	$^{241}\text{Am}$	$^{90}\text{Sr}$	$^{144}\text{Ce}$	$^{137}\text{Cs}$	$^{60}\text{Co}$
78-2-1	(A)	29.99	-0.4±0.4	0.2±0.7	-1.0±1.0	-4±4	-4±4	2.5±1.3	2±1
78-2-1	(B)	29.99	-0.9±0.7	1.2±0.7	-0.57±1.2	3±4	-9±6	3±3	-4±2
78-2-2	(A)	30.33	0.2±0.5	0.3±0.5	-1.0±1.0	2±5	-13±6	-2±3	-2±3
78-2-2	(B)	30.33	0.1±0.5	0.9±0.8	1.0±5.0	1±3	-10±6	2±3	-5±3
78-2-3	(A)	30.54	0.1±0.5	0.2±0.5	-0.3±0.9	3±4	-5±6	-1±2	-3±2
78-2-3	(B)	30.54	0.2±0.5	0.9±0.9	0.1±0.1	4±3	-4±6	-5±2	-4±2
78-2-4	(A)	31.58	-0.6±0.3	0.6±0.4	0.0043±0.8	0±3	-3±6	-3±2	-2±2
78-2-4	(B)	31.58	0.7±0.8	2.0±1.0	1.0±0.9	2±3	-17±6	-2±2	-4±2
78-2-5	(A)	70.65	-0.9±0.8	0.5±0.6	-1.0±1.0	4±4	10±10	4±2	-3±2
78-2-5	(B)	70.65	-0.034±0.57	-0.4±0.7	-0.26±1.3	0±5	-10±10	1±2	-7±2
78-2-6	(A)	71.84	2.0±2.0	-0.1±0.3	33±3.0 [a]	2±3	8±14	-1±2	-1±2
78-2-6	(B)	71.84	0.4±0.7	0.3±0.9	-0.31±1.3	-7±4	-5±6	2±2	-3±2
78-2-7	(A)	72.66	-0.013±0.48	-0.026±0.49	0.4±0.7	3±4	-6±6	-6±3	-3±2
78-2-7	(B)	72.66	0.2±0.8	-0.7±0.5	0.7±0.9	4±4	-2±8	-10±4	-2±3
78-2-8	(A)	74.10	-0.8±0.4	-0.2±0.4	10.0±10.0	3±4	-10±10	3±2	-6±3
78-2-8	(B)	74.10	0.6±0.7	0.1±0.7	0.9±10.0	-3±5	0±9	-13±4	-3±3
78-3-1	(A)	28.10	-0.4±0.5	1.8±0.7 [a]	0.2±1.3	-2±4	-7±6	-3±2	-3±2
78-3-1	(B)	28.10	-0.6±0.4	0.4±0.4	-1.0±1.0	1±4	-12±7	1±2	-5±2
78-3-2	(A)	68.21	0.6±0.6	0.5±0.4	-1.0±1.0	2±4	-1±10	2±2	-3±2
78-3-2	(B)	68.21	-0.8±0.6	0.9±0.7	0.94±1.2	4±3	20±20	0±2	-3±3
78-3-3	(A)	69.16	-0.07±0.32	0.002±0.48	-0.34±1.3	0±3	10±10	0±3	0±3
78-3-3	(B)	69.16	-1.0±0.5	-0.1±0.5	0.48±1.0	-1±4	-5±7	7±2 [a]	-6±3
78-3-4	(A)	70.20	0.9±0.7	0.2±0.2	0.2±0.9	2±3	9±15	4±3	1±3
78-3-4	(B)	70.20	1.2±0.8	-0.059±0.51	-0.53±1.0	4±4	3±7	1±3	-9±3
78-3-5	(A)	71.81	2.0±1.0	0.4±0.6	-0.65±1.3	2±3	-15±8	1±2	2±2
78-3-5	(B)	71.81	2.0±3.0	0.15±1.4	-0.23±1.4	-4±4	4±8	-12±4	-2±3
78-3-6	(A)	72.76	-0.022±0.95	0.2±0.5	0.6±0.7	8±5	-6±7	-1±3	-5±2
78-3-6	(B)	72.76	1.0±1.0	1.3±0.9	0.2±0.8	0±4	-16±7	-0.4±3	-1±3
78-3-7	(A)	73.88	-0.021±0.46	-0.042±0.49	0.9±0.9	2±4	4±8	2±3	-1±3
78-3-7	(B)	73.88	-0.7±0.6	-0.5±0.6	2.0±1.0	12±4 [a]	-8±7	3±3	-3±3
78-5-1	(A)	31.00	3.1±1.0 [a]	0.9±0.9	2.0±1.0	-5±3	3±6	4±2	-2±2
78-5-1	(B)	31.00	1.7±0.8 [a]	0.025±0.77	-1.0±0.7	-5±4	-8±7	3±2	-5±2
78-5-2	(A)	32.37	0.3±0.5	0.4±0.5	0.6±0.9	6±4	-19±7	-1±3	-3±3

### 3-20 Review of the transport of selected radionuclides in the Interim Risk Assessment

Table 3-6. Concentrations of radionuclides  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{90}\text{Sr}$ ,  $^{144}\text{Ce}$ ,  $^{137}\text{Cs}$ , and  $^{60}\text{Co}$  in sediment samples collected in 1978 from wells 78-2, 78-3, and 78-5 (from Humphrey, 1980; Table 1)—Continued

Sample no.	Sample depth	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$	$^{241}\text{Am}$	$^{90}\text{Sr}$	$^{144}\text{Ce}$	$^{137}\text{Cs}$	$^{60}\text{Co}$
78-5-2 (B)	32.37	0.2±0.9	-0.041±0.75	0.5±0.9	-4±4	-3±7	-2±3	-3±3
78-5-3 (A)	33.59	0.13±0.65	0.4±0.3	7.0±4.0	-2±3	-4±14	4±2	4±3
78-5-3 (B)	33.59	0.2±0.5	1.3±0.9	1.0±1.0	5±3	-4±8	-10±4	-3±3
78-5-4 (A)	34.26	-0.2±0.5	0.9±0.5	0.3±0.9	2±3	-6±8	1±2	-4±2
78-5-4 (B)	34.26	-0.059±0.45	1.9±0.9 [a]	0.84±1.1	5±3	-12±6	-1±2	0±2
78-5-5 (A)	69.04	0.048±0.18	1.4±0.9	0.53±2.4	7±4	5±7	0±3	-2±2
78-5-5 (B)	69.04	0.8±0.9	-0.071±0.45	2.0±3.0	2±3	1±11	5±3	-4±3
78-5-6 (A)	70.13	-0.9±0.9	0.7±0.8	-0.91±1.2	-6±4	-26±8	-4±3	1±2
78-5-6 (B)	70.13	0.7±0.7	-0.9±0.6	2.0±1.0	1±4	-20±20	1±2	-5±2
78-5-7 (A)	70.99	-0.9±0.7	0.074±0.5	-0.75±1.5	-1±4	-3±8	5±2 [a]	-3±2
78-5-7 (B)	70.99	-0.049±0.83	-0.7±0.6	0.82±2.2	2±4	1±8	1±2	3±2
78-5-8 (A)	73.24	-0.9±0.7	0.3±0.5	0.44±1.1	8±5	1±6	-3±3	4±3
78-5-8 (B)	73.24	3.0±1.0 [a]	13±2.0 [a]	3.0±4.0	-1±4	10±20	4±3	3±3